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Influence of an External Magnetic Field on the Reactions of Transition Metal Energetic Complexes

by Jennifer L. Gottfried, Jacob Henebry, Shivaiah Vaddypally,
and Michael J. Zdilla

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Influence of an External Magnetic Field on the Reactions of Transition Metal Energetic Complexes

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1. Introduction

The influence of magnetic fields on plasmas is an important area of research in fields such as astrophysics, fusion research, analytical chemistry, propulsion of space vehicles, and other military applications. Direct modification of burn rates through applied electromagnetic fields offers the potential for tailored, on-demand throttle control or remote countermeasures for adverse combustion phenomena. Magnetic fields have been shown to influence burn rates, shock velocities, and deflagration to detonation transitions.¹⁻¹⁰

In the U.S. Army Combat Capabilities Development Command (DEVCOM) Army Research Laboratory (ARL), we developed magnetic-field-enhanced laser-induced air shock from energetic materials (B-LASEM) to investigate the influence of a magnetic field on the energy release of milligram quantities of material. This system has been used to study the laser-initiated, microsecond-timescale energy release and combustion of metal plates,¹¹ metal powders,¹¹⁻¹² conventional military explosives,¹³ magnet-in-ferroelectric energetic crystals,¹⁴ and chiral molecular ferroelectric energetic crystals.¹⁵

The effect of a 0.78-T magnetic field on the energy release of aluminum (Al), copper (Cu), or iron (Fe) powders (and mixtures of Al+Cu or Al+Fe) during three time regimes was previously investigated.¹² The magnetic field was shown to enhance emission intensities and plume expansion rates during the laser-induced plasma ($<10\ \mu\text{s}$), the post-plasma combustion ($\sim 10\text{--}500\ \mu\text{s}$), and self-sustained combustion regimes ($\sim 1\text{--}100\ \text{ms}$). The plasma temperatures and electron densities were also increased, regardless of the magnetic properties of the materials. In contrast, the observed differences in combustion intensities and velocities were dependent on the magnetic properties. No significant differences based on magnetic field direction were observed.

For example, the results from the high-speed schlieren imaging for Fe powder are shown in Figure 1. Both the plume size and luminosity grow with increasing strength of the applied magnetic field (where $7800\ \text{G} = 0.78\ \text{T}$). Emission intensities from the laser-induced plasma spectra increased for the metal emission features, but decreased for the organic species resulting from contaminants, surface functional groups, or entrained air (Figure 2). Both the post-plasma combustion plume velocities and IR emission from the self-sustained combustion reactions also increased in the magnetic field (Figure 3). The characteristic laser-induced shock velocities (early single-digit microsecond-timescale energy release) and Dewey shock velocities (energy release up to $13\ \mu\text{s}$) decreased for Fe under the magnetic field, while they increased or did not change for Al and Cu.

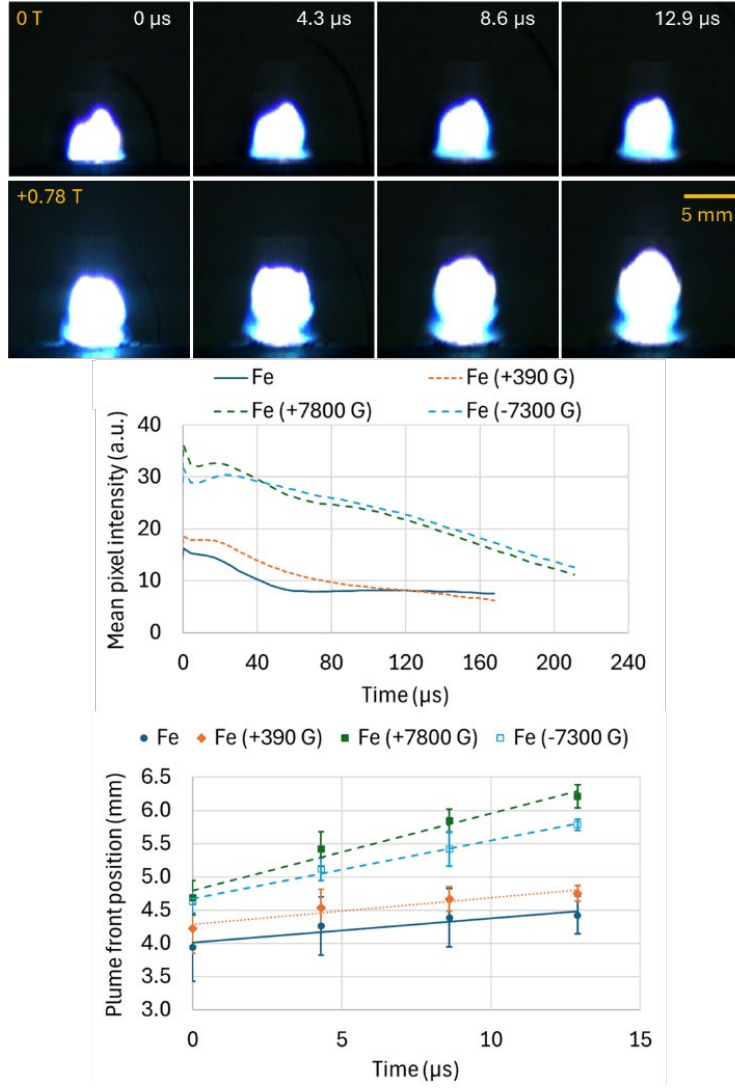


Figure 1. Snapshots from the high-speed schlieren videos of $<10\text{-}\mu\text{m}$ Fe (99.5%) powder (top), the mean pixel intensity in each frame (middle), and the plume front position in each frame relative to the sample surface (bottom).¹²

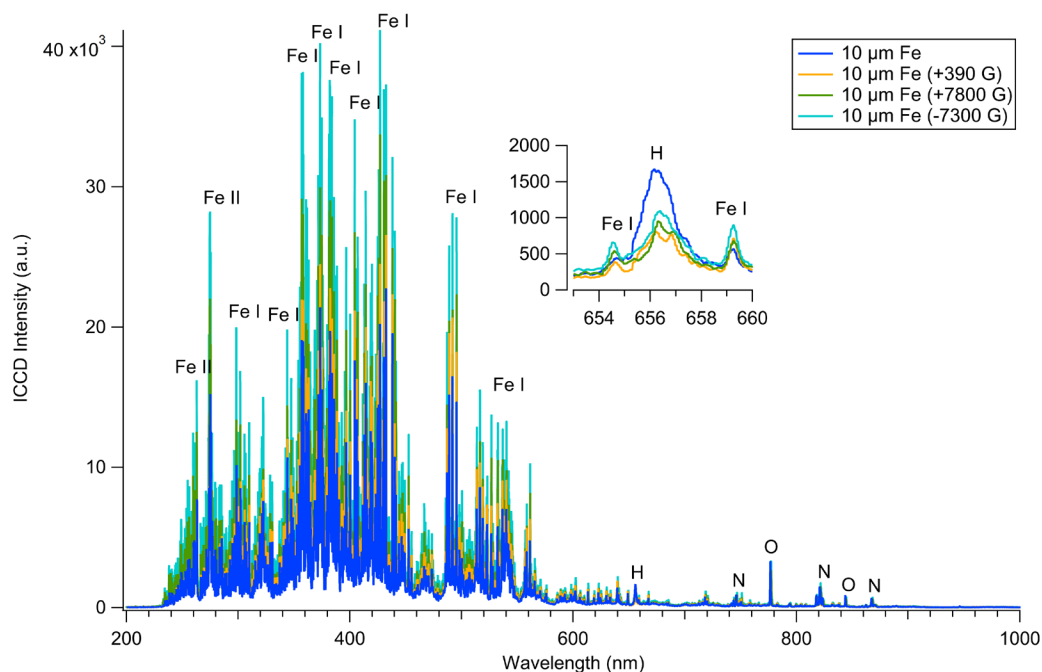


Figure 2. Laser-induced plasma emission spectra for <10-μm Fe (99.5%) powder with and without the presence of an external magnetic field.¹²

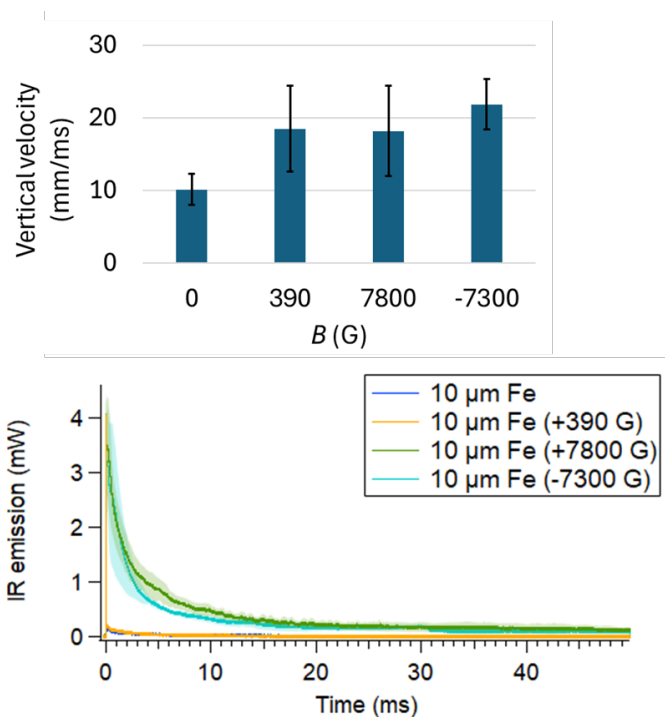


Figure 3. Post-plasma combustion plume front velocities (top) and IR emission from self-sustained combustion reactions (bottom) for <10-μm Fe (99.5%) powder with and without the presence of an external magnetic field.¹²

Recently, a new class of redox-frustrated, high-energy-density materials based on transition metal complexes has been developed. The magnetic ordering of the

transition metal ions influence the pathway and rates of decomposition. It was demonstrated that a 0.5-T magnetic field modulated the ignition onset temperature of $\text{Mn}_6(\mu_3\text{NTz}'\text{BuMe})_6(\text{ClO}_4)_6$ by +10.4 °C.¹⁶ Here, the energy release behavior of energetic complexes based on manganese (Mn), Fe, cobalt (Co), nickel (Ni), Cu, zinc (Zn) or sodium (Na) with or without an 0.78-T magnetic field was compared using B-LASEM.

2. Experimental

2.1 Samples

The samples investigated in this work comprise a series of transition metal-based nitrotetrazole (NO_2Tz) ligands and a Mn-based cluster of Mn^{III} and Mn^{IV} ions with 3,6-dinitrobenzoate (DNOBz) ligands that have an oxygen balance similar to the explosive TNT. The list of sample abbreviations and molecular formulae is shown below:

- $\text{NaNO}_2\text{Tz} = \text{Na}(\text{NO}_2\text{Tz})(\text{H}_2\text{O})_2$
- $\text{CuNO}_2\text{Tz} = [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Cu}(\text{NO}_2\text{Tz})_3\}_n]$
- $\text{CoNO}_2\text{Tz} = [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Co}(\text{NO}_2\text{Tz})_3\}_n]$
- $\text{FeNO}_2\text{Tz} = [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Fe}(\text{NO}_2\text{Tz})_3\}_n]$
- $\text{Mn12} = [\text{Mn}_{12}\text{O}_{12}(\text{DNOBz})_{16}(\text{H}_2\text{O})_2(\text{CH}_3\text{CN})_2]$
- $\text{MnNO}_2\text{Tz}: [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Fe}(\text{NO}_2\text{Tz})_3\}_n]$
- $\text{NiNO}_2\text{Tz}: [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Ni}(\text{NO}_2\text{Tz})_3\}_n]$
- $\text{ZnNO}_2\text{Tz}: [\text{Na}(\text{H}_2\text{O})_3]_n[\{\text{Zn}(\text{NO}_2\text{Tz})_3\}_n]$

Figure 4 shows the crystal structure of the Mn12 cluster.

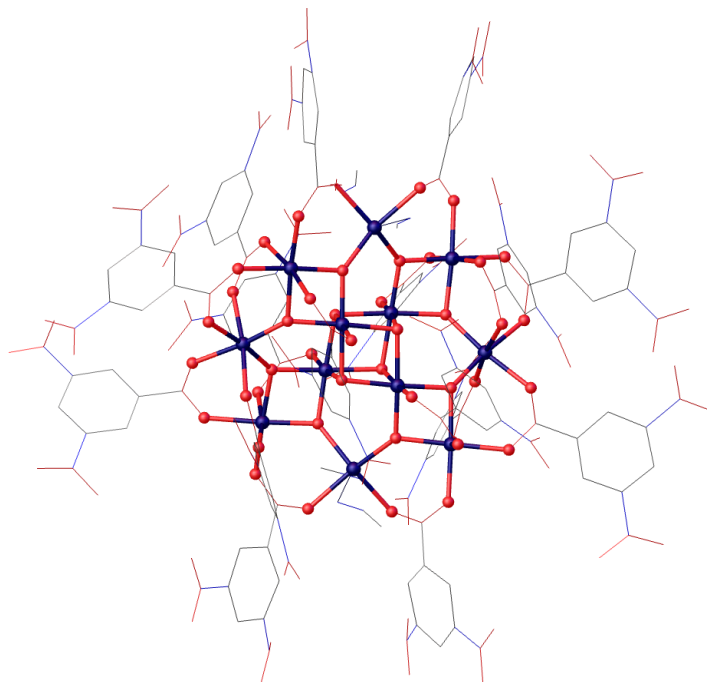


Figure 4. Crystal structure of the Mn₁₂ cluster. Nitrophenyl groups (except for ligating oxygen) shown in wire mode for clarity.

Sample names annotated with “P” correspond to data obtained under a +0.78-T magnetic field, while names annotated with “N” correspond to a –0.73-T field (with the field direction defined as discussed in the next section). Samples were prepared by pressing 10–15 mg of powder into double-sided tape affixed to a glass microscope slide.

2.2 B-LASEM

The B-LASEM system used in this work¹² (Figure 5) is a modification of the LASEM-420 system previously described in detail⁴ and a previous version of B-LASEM that incorporated a weak electromagnet (390 G).¹¹ To investigate the influence of a magnetic field on the energy release of laser-excited materials in three distinct time regimes ($<10\ \mu\text{s}$, $\sim 10\text{--}500\ \mu\text{s}$, and $\sim 1\text{--}100\ \text{ms}$), the sample was placed on a permanent magnet setup consisting of two neodymium permanent magnets (50.8 mm [2 inch] diameter, 12.7 mm [0.5 inch] thick, N52 grade) stacked and rotated such that the sample was placed on the edge of the magnet at the precise locations with the strongest magnetic field as measured with a transverse Hall probe (FW Bell 5170 Gauss/Tesla Meter), as shown in Figure 6. The magnetic flux lines exit the north pole (positive magnetic-flux density B) and enter the south pole (negative B); thus, the electrons flow in opposite directions parallel to the sample surface depending on which pole the sample is above. A map of the measured B

across the magnet edge is shown in Figure 7, while the decrease in B in the air above the sample surface (where the laser-initiated chemical reactions occur) is shown in Figure 8.

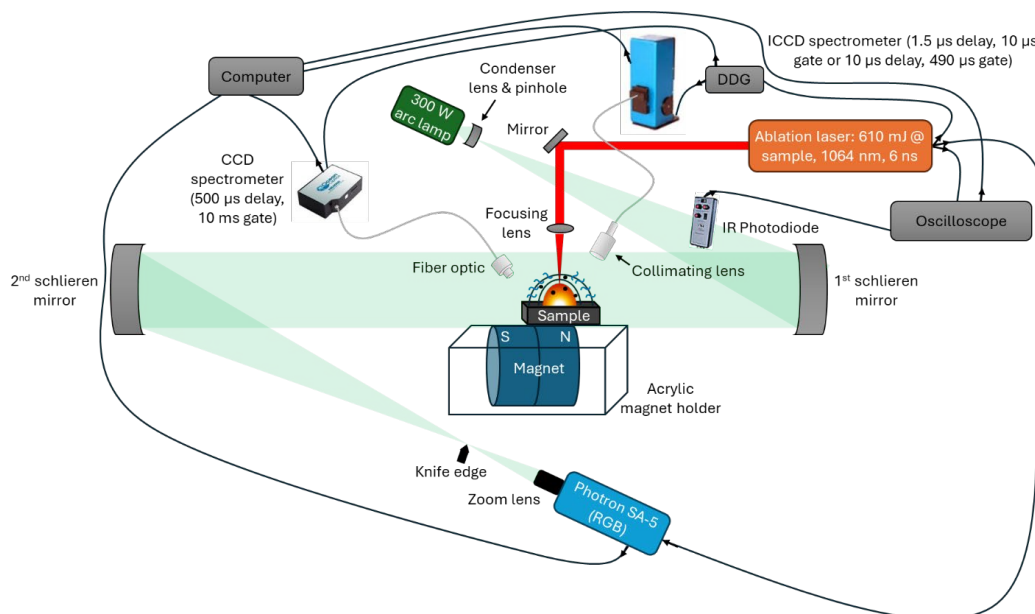


Figure 5. Experimental schematic for the B-LASEM system as configured for the current study; figure is not to scale.

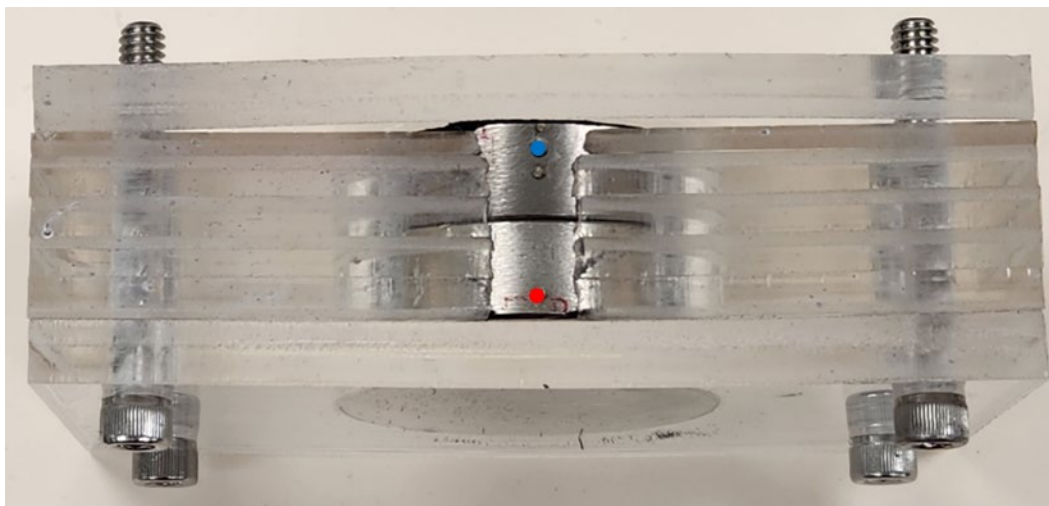


Figure 6. Top-down view of the stacked permanent magnets affixed within an acrylic holder. The red and blue dots correspond to the locations of the maximum positive and negative magnetic-flux densities, respectively.

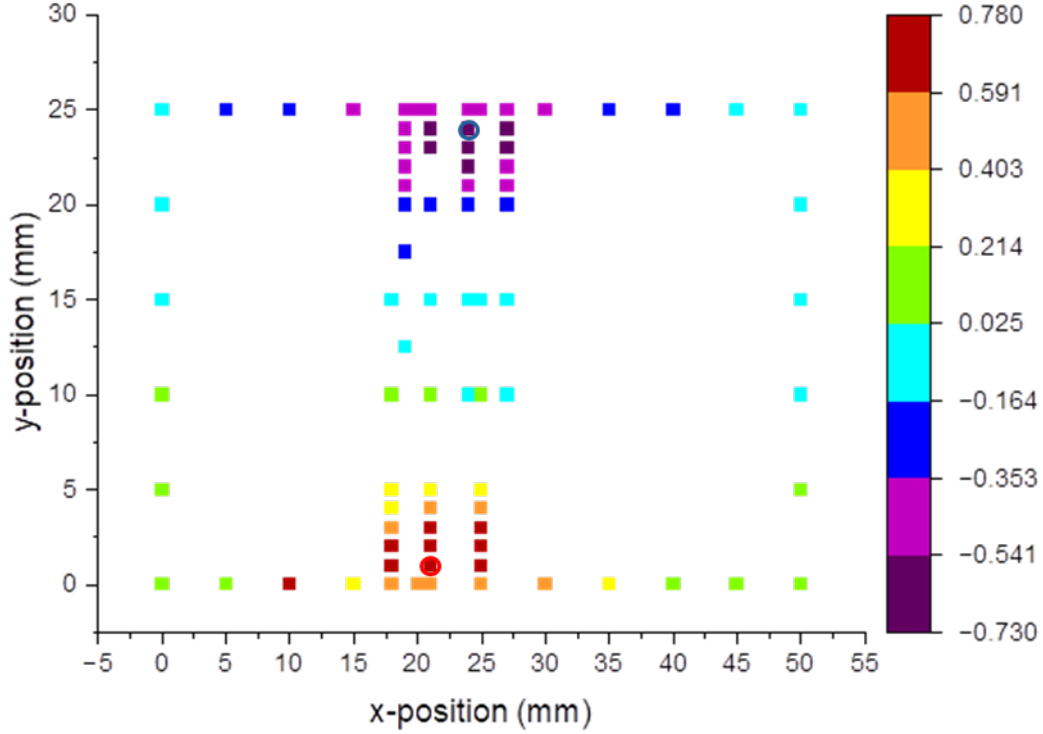


Figure 7. Map of the measured magnetic-flux densities (color scale is in T) across the edge surface of the bare stacked magnets. The red and blue outlined circles correspond to the laser focal spots marked in Figure 2.

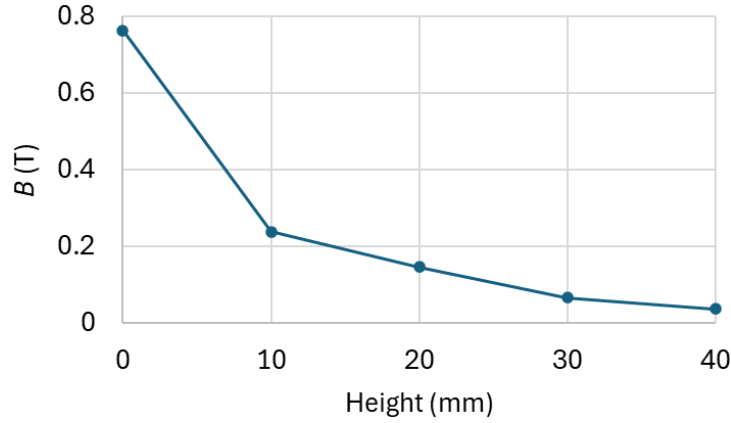


Figure 8. Magnetic-flux density B as a function of the height above the surface of the magnet edge.

A high-energy nanosecond-pulsed laser is focused just below the sample surface to ablate, ionize, and excite the material into a laser-induced plasma that emits radiation at wavelengths characteristic of the species reacting in the plasma. A laser-induced shock wave expands into the air above the sample surface, and particles are ejected off the sample surface to react during post-plasma combustion reactions. High-speed schlieren imaging with a color camera (red–green–blue [RGB]) is used to record the expansion of the shock wave, plasma plume, and

subsequent combustion reactions; the 4.25-inch-diameter schlieren mirrors have a 45-inch focal length and are separated by 90 inches. High-speed schlieren videos (20 replicates each) were obtained at 420 kfps with an image size of 832×16 pixels and an exposure time of 369 ns to track the expansion of the laser-induced shock wave into the air above the samples up to 112 μ s. Custom MATLAB code was used to measure the time-resolved shock wave positions¹⁷⁻¹⁸; for these measurements the shock position was rounded down to the nearest pixel.¹⁹ The code also automatically fits the data to determine the extent of energy release on the microsecond timescale using parameters such as the characteristic laser-induced shock velocity, the Dewey shock velocity, and the speed of sound in air for the acoustic wave.²⁰ For the current samples, no significant differences in calculated parameters were observed when the shock position was rounded up to the next pixel. Additional custom code was used to perform multiple comparison of means tests on the resulting calculated parameters to determine the statistical significance of the differences in the average parameters for each sample.²¹

Two spectrometers monitored the spatially and temporally integrated plasma or combustion emission. A digital delay generator (DDG) controlled the gate delays and gate widths for the spectrometers; the Q-switch out trigger from the laser initiated all data collection. A high-resolution echelle spectrometer with an intensified charge-coupled device (ICCD) detector resolved the spatially and temporally integrated emission from the post-plasma combustion (gate delay = 10 μ s, gate width = 490 μ s) or laser-induced plasma (gate delay set to 1.5 μ s to avoid the continuum emission, gate width set to 10 μ s over the plasma lifetime). The post-plasma combustion emission from the first 10 laser shots was saved; the second set of 10 laser shots was used to record the laser-induced plasma emission. The ICCD spectrometer was wavelength-calibrated with a mercury lamp and intensity-calibrated with a deuterium lamp. A charge-coupled device (CCD) spectrometer was set to record the combustion emission spectra from 500 μ s to 10.5 ms to focus on the self-sustained combustion reactions. An IR-sensitive photodiode (90–1700 nm) was used to monitor the time-resolved emission from self-sustained combustion on the millisecond timescale; the reported signals from the photodiode were corrected for the instrument gain (1×10^3) and converted to optical power. The first five laser shots were widely spaced along the sample surface to ensure sufficient ejected particles for the self-sustained combustion reactions; the IR traces and CCD spectra for these first five shots were saved for each sample. Subsequent laser shots (for which only the shock data and ICCD spectra were saved) were more closely spaced since that data does not significantly depend on the amount of intact particles ejected from the surface by the shock wave. All error bars in the figures represent 95% confidence intervals (CIs).

3. Results

3.1 High-Speed Schlieren Imaging of Laser-Induced Shock Waves

Snapshots from the high-speed schlieren videos for the Mn-based samples with and without the magnetic field are shown in Figures 9 and 10; snapshots from the remaining samples are shown in Appendix A. The images are rotated 90° (with the sample on the far left and the direction of laser propagation from right to left), cropped, and the brightness enhanced by 80%; the scale bar applies to all images. Each vertically stacked frame corresponds to 2.38 μs (with a 369 ns shutter). Emission from the laser-induced plasma and subsequent combustion reactions are visible in the images, as is the primary shock front (plus several weaker shock fronts²²) and the change in refractive index resulting from expansion of the hot gaseous products in the reaction region. Mean pixel intensities averaged across each frame for each laser shot were calculated for each sample to quantitatively compare the visible emission; the contribution of the schlieren light source to the mean pixel intensity has been background-subtracted for each video using the value for the frame prior to the laser firing. The visible emission and reaction plume height are two complementary indicators of combustion reactions since not all combustion species produce visible emissions.

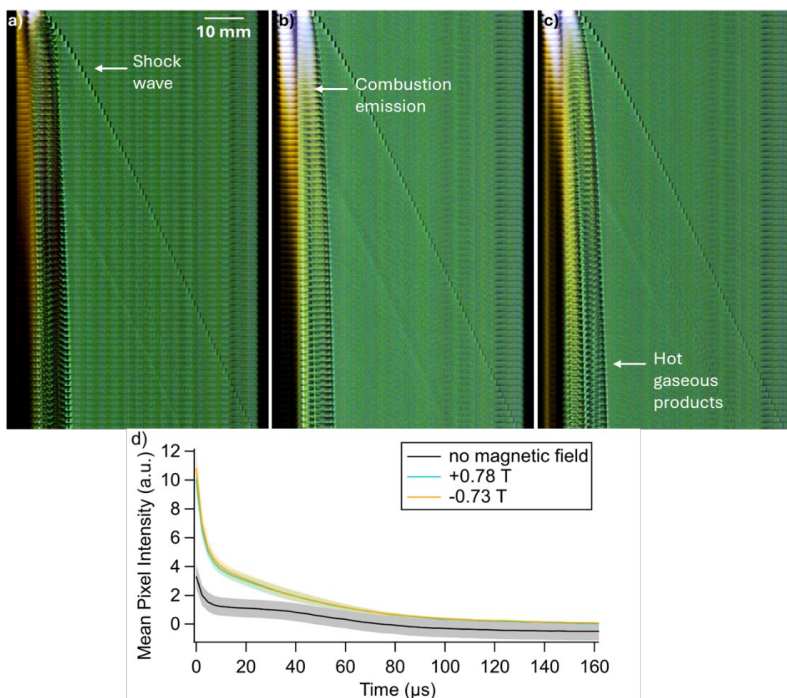


Figure 9. Snapshots from representative high-speed schlieren videos of the a) MnNO₂Tz, b) MnNO₂Tz (+0.78 T), and c) MnNO₂Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

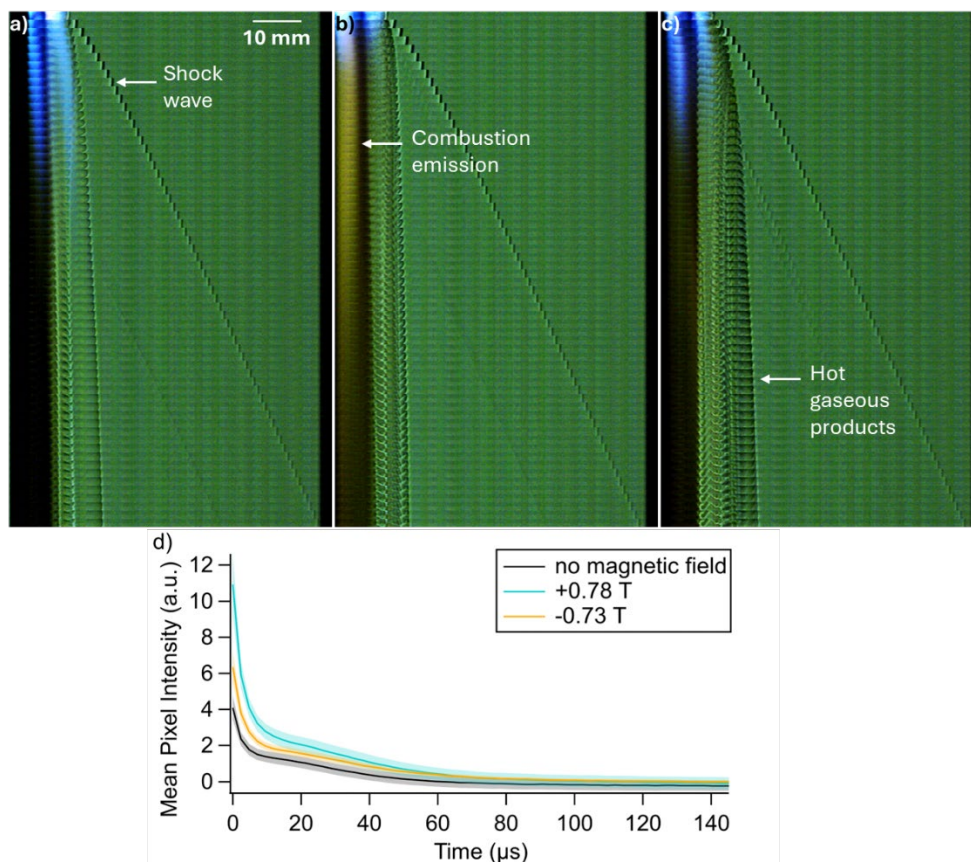


Figure 10. Snapshots from representative high-speed schlieren videos of the a) Mn12, b) Mn12 (+0.78 T), and c) Mn12 (−0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

The combustion emission for the NaNO_2Tz is stronger and longer-lasting in the presence of the magnetic field (peaking around 30 μs , Appendix A, Figure A-1), while the plume of expanding gases is shorter at 145 μs (Figure 11). While the emission for the CuNO_2Tz is also stronger in the magnetic field, its duration is comparable (see Figure A-2) and the average height of the plume of expanding gases is slightly shorter. The CoNO_2Tz combustion intensity is stronger and longer-lasting in the magnetic field (see Figure A-3) and the plume of hot gases expands faster—with significant shot-to-shot variation (Figure 11). For FeNO_2Tz , the combustion lasts longer in the magnetic field (see Figure A-4) but the plume of expanding gases is significantly contracted. The plasma emission for MnNO_2Tz lasts noticeably longer in the magnetic field (see Figure 9) and the reaction plumes are larger, while the magnetic field has a significantly lessened effect on the emission Mn12 sample (see Figure 10) even as the plume heights decrease significantly (Figure 11). The plasma emission for MnNO_2Tz is more white/orange than for Mn12 (which is blue/purple)—this is because of the large Na content present in the MnNO_2Tz , whose emission near 589 nm obscures the Mn emission

at lower wavelengths (Figure 12). The magnetic field has a smaller effect on the combustion emission from NiNO_2Tz (see Figure A-5), though the plume heights decrease. In contrast, the early time post-plasma combustion for ZnNO_2Tz is significantly enhanced by the magnetic field (see Figure A-6) while no change in plume height occurs (Figure 11).

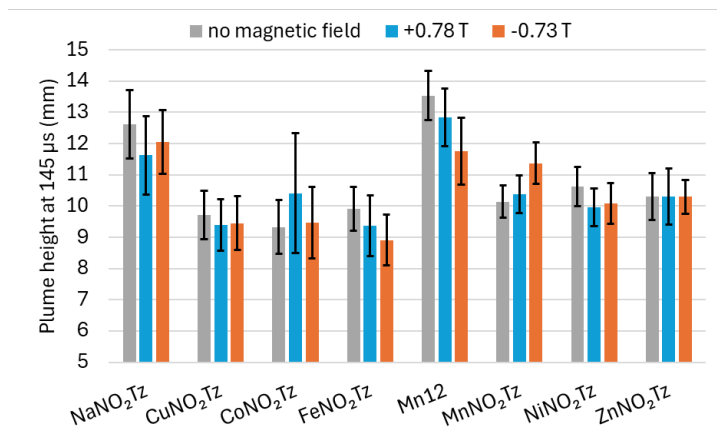


Figure 11. Measured plume heights at 145 μs for each sample with and without the magnetic field; error bars are 95% CIs.

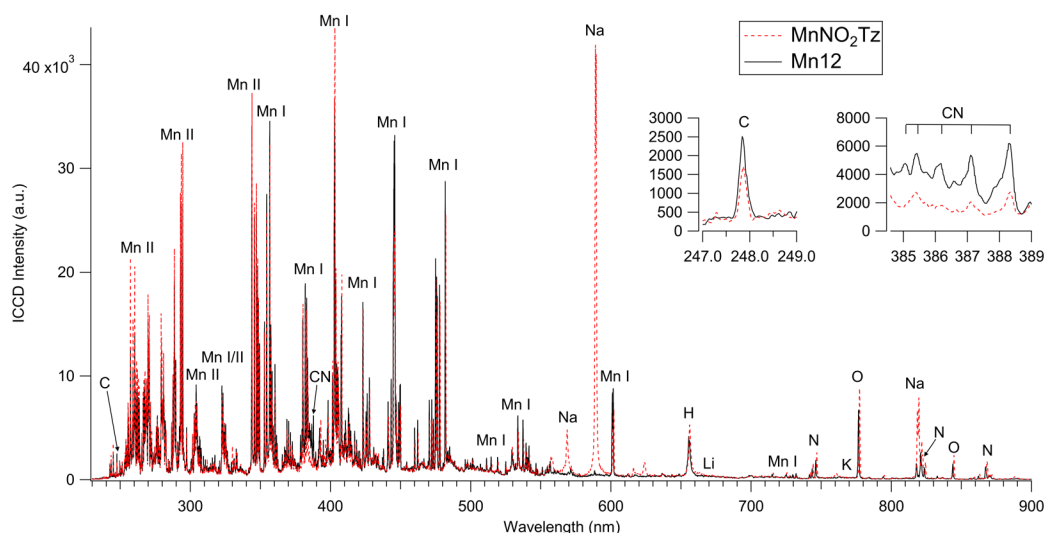


Figure 12. Comparison of the laser-induced plasma spectra for MnNO_2Tz and Mn12 .

After the laser-induced shock front positions in each video frame were obtained by the custom MATLAB code, the shock wave velocity versus time was fit to various functional forms to determine the extent of energy release on different timescales.²³ The characteristic laser-induced shock velocities for each laser shot were determined as the y-intercepts to a 5th order fit of the shock velocity data; these values quantify the energy release on the early single-digit microsecond timescale

and, for detonable materials, directly correlate to measured detonation velocities from large-scale testing.²⁴ The shock velocities at 13 μ s from a fit of the shock velocity data to the Dewey equation (empirically determined from TNT blast data) quantify the total energy release in the laser-induced plasma. The correlation between the averages of the two parameters for the current samples are shown in Figure 13 along with the 95% CIs; the presented values are relative to those of NaNO_2Tz —the absolute values fall below those of conventional military explosives but higher than inert materials. Generally, the characteristic laser-induced shock velocities and Dewey shock velocities are linearly correlated; however, energetic materials that react later in the plasma lifetime (e.g., because of slower-reacting ingredients) show greater increases in Dewey shock velocity than characteristic laser-induced shock velocities (i.e., fall below the expected linear trendline)—while faster-reacting materials fall above the linear trendline.

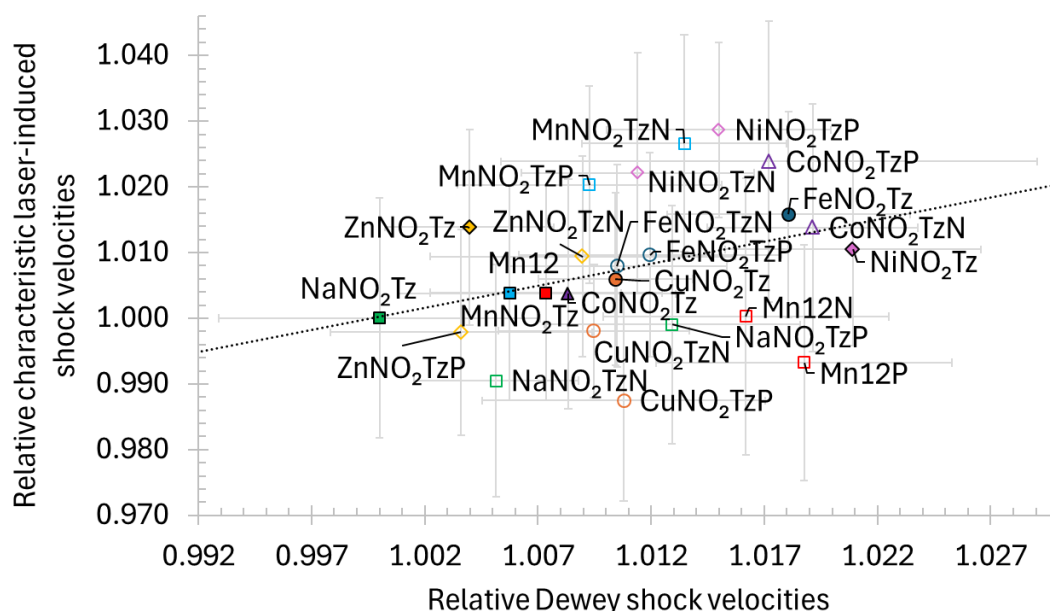


Figure 13. Comparison of the normalized characteristic laser-induced shock velocities and Dewey shock velocities for the NO_2Tz and Mn12 samples with (open symbols) and without (solid symbols) the external magnetic field. The dotted line shows a linear fit to the data to guide the eye and the error bars represent 95% CIs.

The trends in characteristic laser-induced shock velocities and Dewey shock velocities under the magnetic field are summarized in Table 1. The CoNO_2Tz and MnNO_2Tz are the only samples to show an increase in both the fast energy release (characteristic laser-induced shock velocities) and the total energy release (Dewey shock velocities) in the laser-induced plasma under the external magnetic field (Table 1); this suggests the combustion is enhanced throughout the single-digit microsecond timescale. They are also the only two samples to show an increase in reaction plume height within the magnetic field (see Figure 11). The FeNO_2Tz is

the only sample to show a decrease in both energy release parameters. CuNO_2Tz and ZnNO_2Tz show a decrease in fast energy release in the magnetic field but no significant change in total energy release (suggesting that the energy release is just shifted until later in the plasma lifetime); both samples showed little or no change in reaction plume heights under the magnetic field. NaNO_2Tz and Mn12 show a decrease in fast energy release and an increase in total energy release—suggesting later timescale combustion reactions more than compensated for the decrease in fast energy release. Finally, NiNO_2Tz showed an increase in fast energy release but a decrease in total energy release, suggesting little or no later timescale combustion.

Table 1. Observed trends in early and total energy release in the laser-induced plasma under the influence of the external magnetic field.

Sample	Early μs energy release	Total energy release
NaNO_2Tz	↓	↑
CoNO_2Tz	↑	↑
CuNO_2Tz	↓	...
FeNO_2Tz	↓	↓
MnNO_2Tz	↓	↑
MnNO_2Tz	↑	↑
NiNO_2Tz	↑	↓
ZnNO_2Tz	↓	...

Because of the significant overlap in 95% CIs for the shock velocities, analysis of variance (ANOVA) testing followed multiple comparison of means testing was performed on the complete dataset to determine what parameters were statistically different.²¹ Many of the mean characteristic laser-induced shock velocities (Table 2) and Dewey shock velocities (Table 3) for the samples are not statistically distinguishable at the 95% confidence level (p-values <0.05), but the FeNO_2Tz and NaNO_2Tz characteristic laser-induced shock velocities are the most indistinguishable with or without the magnetic field (i.e., they have greater than 40% probabilities that the difference in means would be incorrectly found significant). The same is true for the Dewey shock velocities of CuNO_2Tz , FeNO_2Tz , and NiNO_2Tz (Table 3). None of the eight samples are distinguishable at the 95% confidence level without the magnetic field, although statistically significant differences are observed in the presence of the magnetic field. While the CIs for the measurements could be reduced by obtaining more laser shots, the similarities in the energy content of the samples without the magnetic field make distinguishing them based on shock velocities challenging. Thus, supporting diagnostic data is required.

Table 2. Multiple comparison of means test results for the characteristic laser-induced shock velocities; p-values <0.05 are shaded in dark red and indicate the average values are different at the 95% confidence level, while p-values shaded in green show unlikely statistical difference.

	CoNO ₂ TzN	CoNO ₂ TzP	CoNO ₂ Tz	CuNO ₂ TzN	CuNO ₂ TzP	CuNO ₂ Tz	FeNO ₂ TzN	FeNO ₂ TzP	FeNO ₂ Tz	Mn12N	Mn12P	Mn12	MnNO ₂ TzN	MnNO ₂ TzP	MnNO ₂ Tz	NaNO ₂ TzN	NaNO ₂ TzP	NaNO ₂ Tz	NiNO ₂ TzN	NiNO ₂ TzP	NiNO ₂ Tz	ZnNO ₂ TzN	ZnNO ₂ TzP	ZnNO ₂ Tz
CoNO ₂ TzN		0.271	0.405	0.195	0.030	0.600	0.630	0.733	0.867	0.438	0.090	0.414	0.290	0.588	0.411	0.055	0.223	0.255	0.490	0.219	0.783	0.717	0.190	0.995
CoNO ₂ TzP	0.271		0.055	0.018	0.001	0.560	0.115	0.151	0.350	0.062	0.006	0.057	0.956	0.571	0.056	0.003	0.022	0.026	0.675	0.909	0.166	0.145	0.017	0.274
CoNO ₂ Tz	0.405	0.055		0.642	0.180	0.175	0.726	0.623	0.318	0.954	0.387	0.988	0.059	0.170	0.993	0.276	0.699	0.759	0.128	0.040	0.571	0.639	0.631	0.402
CuNO ₂ TzN	0.195	0.018	0.642		0.381	0.069	0.415	0.339	0.143	0.601	0.690	0.631	0.019	0.066	0.635	0.532	0.937	0.874	0.047	0.012	0.299	0.350	0.988	0.193
CuNO ₂ TzP	0.030	0.001	0.180	0.381		0.007	0.091	0.067	0.020	0.162	0.633	0.175	0.001	0.007	0.177	0.802	0.339	0.301	0.004	0.001	0.055	0.071	0.389	0.030
CuNO ₂ Tz	0.600	0.560	0.175	0.069	0.007		0.314	0.387	0.721	0.194	0.027	0.180	0.593	0.987	0.178	0.015	0.082	0.097	0.869	0.480	0.420	0.375	0.067	0.604
FeNO ₂ TzN	0.630	0.115	0.726	0.415	0.091	0.314		0.888	0.516	0.769	0.225	0.737	0.124	0.306	0.733	0.150	0.462	0.512	0.241	0.087	0.831	0.905	0.406	0.625
FeNO ₂ TzP	0.733	0.151	0.623	0.339	0.067	0.387	0.888		0.611	0.664	0.176	0.634	0.162	0.378	0.630	0.114	0.381	0.425	0.303	0.116	0.944	0.983	0.331	0.728
FeNO ₂ Tz	0.867	0.350	0.318	0.143	0.020	0.721	0.516	0.611		0.346	0.063	0.325	0.373	0.709	0.322	0.037	0.166	0.192	0.601	0.288	0.656	0.596	0.139	0.872
Mn12N	0.438	0.062	0.954	0.601	0.162	0.194	0.769	0.664	0.346		0.357	0.966	0.067	0.188	0.962	0.251	0.658	0.716	0.143	0.045	0.610	0.680	0.591	0.435
Mn12P	0.090	0.006	0.387	0.690	0.633	0.027	0.225	0.176	0.063	0.357		0.379	0.006	0.026	0.382	0.821	0.632	0.577	0.017	0.004	0.150	0.183	0.701	0.089
Mn12	0.414	0.057	0.988	0.631	0.175	0.180	0.737	0.634	0.325	0.966	0.379		0.061	0.175	0.996	0.269	0.689	0.748	0.132	0.041	0.581	0.649	0.620	0.410
MnNO ₂ TzN	0.290	0.956	0.059	0.019	0.001	0.593	0.124	0.162	0.373	0.067	0.006	0.061		0.605	0.060	0.003	0.023	0.028	0.712	0.864	0.178	0.155	0.018	0.293
MnNO ₂ TzP	0.588	0.571	0.170	0.066	0.007	0.987	0.306	0.378	0.709	0.188	0.026	0.175	0.605		0.173	0.014	0.079	0.093	0.881	0.490	0.411	0.366	0.064	0.593
MnNO ₂ Tz	0.411	0.056	0.993	0.635	0.177	0.178	0.733	0.630	0.322	0.962	0.382	0.996	0.060	0.173		0.272	0.693	0.752	0.131	0.040	0.577	0.645	0.624	0.407
NaNO ₂ TzN	0.055	0.003	0.276	0.532	0.802	0.015	0.150	0.114	0.037	0.251	0.821	0.269	0.003	0.014	0.272		0.481	0.433	0.009	0.002	0.095	0.119	0.542	0.054
NaNO ₂ TzP	0.223	0.022	0.699	0.937	0.339	0.082	0.462	0.381	0.166	0.658	0.632	0.689	0.023	0.079	0.693	0.481		0.936	0.057	0.015	0.338	0.392	0.925	0.221
NaNO ₂ Tz	0.255	0.026	0.759	0.874	0.301	0.097	0.512	0.425	0.192	0.716	0.577	0.748	0.028	0.093	0.752	0.433	0.936		0.068	0.018	0.381	0.438	0.861	0.253
NiNO ₂ TzN	0.490	0.675	0.128	0.047	0.004	0.869	0.241	0.303	0.601	0.143	0.017	0.132	0.712	0.881	0.131	0.009	0.057	0.068		0.589	0.330	0.293	0.046	0.494
NiNO ₂ TzP	0.219	0.909	0.040	0.012	0.001	0.480	0.087	0.116	0.288	0.045	0.004	0.041	0.864	0.490	0.040	0.002	0.015	0.018	0.589		0.129	0.111	0.011	0.221
NiNO ₂ Tz	0.783	0.166	0.571	0.299	0.055	0.420	0.831	0.944	0.656	0.610	0.150	0.581	0.178	0.411	0.577	0.095	0.338	0.381	0.330	0.129		0.927	0.292	0.778
ZnNO ₂ TzN	0.717	0.145	0.639	0.350	0.071	0.375	0.905	0.983	0.596	0.680	0.183	0.649	0.155	0.366	0.645	0.119	0.392	0.438	0.293	0.111	0.927		0.342	0.712
ZnNO ₂ TzP	0.190	0.017	0.631	0.988	0.389	0.067	0.406	0.331	0.139	0.591	0.701	0.620	0.018	0.064	0.624	0.542	0.925	0.861	0.046	0.011	0.292	0.342		0.188
ZnNO ₂ Tz	0.995	0.274	0.402	0.193	0.030	0.604	0.625	0.728	0.872	0.435	0.089	0.410	0.293	0.593	0.407	0.054	0.221	0.253	0.494	0.221	0.778	0.712	0.188	

Table 3. Multiple comparison of means test results for the Dewey shock velocities; p-values <0.05 are shaded in dark red and indicate the average values are different at the 95% confidence level, while p-values shaded in green show unlikely statistical difference.

	CoNO ₂ TzN	CoNO ₂ TzP	CoNO ₂ Tz	CuNO ₂ TzN	CuNO ₂ TzP	CuNO ₂ Tz	FeNO ₂ TzN	FeNO ₂ TzP	FeNO ₂ Tz	Mn12N	Mn12P	Mn12	MnNO ₂ TzN	MnNO ₂ TzP	MnNO ₂ Tz	NaNO ₂ TzN	NaNO ₂ TzP	NaNO ₂ Tz	NiNO ₂ TzN	NiNO ₂ TzP	NiNO ₂ Tz	ZnNO ₂ TzN	ZnNO ₂ TzP	ZnNO ₂ Tz
CoNO ₂ TzN		0.100	0.219	0.411	0.416	0.111	0.598	0.717	0.890	0.161	0.549	0.038	0.263	0.614	0.244	0.136	0.217	0.021	0.227	0.645	0.353	0.685	0.169	0.180
CoNO ₂ TzP	0.100		0.004	0.014	0.015	0.001	0.030	0.045	0.131	0.791	0.290	0.000	0.587	0.032	0.005	0.002	0.004	0.000	0.648	0.233	0.452	0.041	0.003	0.003
CoNO ₂ Tz	0.219	0.004		0.683	0.677	0.714	0.483	0.386	0.172	0.009	0.068	0.396	0.019	0.468	0.950	0.793	0.995	0.277	0.015	0.091	0.030	0.411	0.883	0.910
CuNO ₂ TzN	0.411	0.014	0.683		0.993	0.439	0.769	0.646	0.337	0.027	0.156	0.209	0.053	0.751	0.730	0.503	0.678	0.136	0.043	0.200	0.079	0.678	0.579	0.603
CuNO ₂ TzP	0.416	0.015	0.677	0.993		0.434	0.776	0.652	0.342	0.027	0.158	0.206	0.054	0.757	0.724	0.498	0.672	0.133	0.044	0.203	0.080	0.684	0.573	0.597
CuNO ₂ Tz	0.111	0.001	0.714	0.439	0.434		0.286	0.218	0.083	0.003	0.029	0.630	0.007	0.275	0.668	0.917	0.719	0.472	0.005	0.040	0.011	0.234	0.826	0.799
FeNO ₂ TzN	0.598	0.030	0.483	0.769	0.776	0.286		0.868	0.505	0.054	0.260	0.122	0.100	0.981	0.523	0.335	0.479	0.074	0.083	0.323	0.144	0.903	0.396	0.416
FeNO ₂ TzP	0.717	0.045	0.386	0.646	0.652	0.218	0.868		0.617	0.078	0.336	0.087	0.138	0.887	0.421	0.259	0.382	0.051	0.117	0.411	0.196	0.965	0.311	0.327
FeNO ₂ Tz	0.890	0.131	0.172	0.337	0.342	0.083	0.505	0.617		0.207	0.645	0.027	0.326	0.521	0.192	0.104	0.170	0.014	0.285	0.747	0.430	0.586	0.130	0.139
Mn12N	0.161	0.791	0.009	0.027	0.027	0.003	0.054	0.078	0.207		0.422	0.001	0.778	0.057	0.010	0.004	0.009	0.000	0.846	0.347	0.624	0.071	0.006	0.006
Mn12P	0.549	0.290	0.068	0.156	0.158	0.029	0.260	0.336	0.645	0.422		0.008	0.602	0.270	0.078	0.037	0.067	0.004	0.543	0.890	0.748	0.315	0.049	0.053
Mn12	0.038	0.000	0.396	0.209	0.206	0.630	0.122	0.087	0.027	0.001	0.008		0.001	0.116	0.362	0.558	0.400	0.812	0.001	0.011	0.003	0.095	0.483	0.462
MnNO ₂ TzN	0.263	0.587	0.019	0.053	0.054	0.007	0.100	0.138	0.326	0.778	0.602	0.001		0.105	0.023	0.009	0.019	0.001	0.930	0.509	0.837	0.127	0.013	0.014
MnNO ₂ TzP	0.614	0.032	0.468	0.751	0.757	0.275	0.981	0.887	0.521	0.057	0.270	0.116	0.105		0.508	0.324	0.464	0.070	0.087	0.335	0.151	0.922	0.383	0.402
MnNO ₂ Tz	0.244	0.005	0.950	0.730	0.724	0.668	0.523	0.421	0.192	0.010	0.078	0.362	0.023	0.508		0.745	0.945	0.251	0.018	0.104	0.035	0.447	0.834	0.861
NaNO ₂ TzN	0.136	0.002	0.793	0.503	0.498	0.917	0.335	0.259	0.104	0.004	0.037	0.558	0.009	0.324	0.745		0.798	0.798	0.007	0.051	0.015	0.278	0.908	0.881
NaNO ₂ TzP	0.217	0.004	0.995	0.678	0.672	0.719	0.479	0.382	0.170	0.009	0.067	0.400	0.019	0.464	0.945	0.798		0.280	0.015	0.090	0.030	0.407	0.889	0.916
NaNO ₂ Tz	0.021	0.000	0.277	0.136	0.133	0.472	0.074	0.051	0.014	0.000	0.004	0.812	0.001	0.070	0.251	0.410	0.280		0.000	0.006	0.001	0.057	0.347	0.330
NiNO ₂ TzN	0.227	0.648	0.015	0.043	0.044	0.005	0.083	0.117	0.285	0.846	0.543	0.001	0.930	0.087	0.018	0.007	0.015	0.000		0.455	0.768	0.107	0.010	0.011
NiNO ₂ TzP	0.645	0.233	0.091	0.200	0.203	0.040	0.323	0.411	0.747	0.347	0.890	0.011	0.509	0.335	0.104	0.051	0.090	0.006	0.455		0.644	0.386	0.067	0.072
NiNO ₂ Tz	0.353	0.452	0.030	0.079	0.080	0.011	0.144	0.196	0.430	0.624	0.748	0.003	0.837	0.151	0.035	0.015	0.030	0.001	0.768	0.644		0.181	0.021	0.023
ZnNO ₂ TzN	0.685	0.041	0.411	0.678	0.684	0.234	0.903	0.965	0.586	0.071	0.315	0.095	0.127	0.922	0.447	0.278	0.407	0.057	0.107	0.386	0.181		0.332	0.350
ZnNO ₂ TzP	0.169	0.003	0.883	0.579	0.573	0.826	0.396	0.311	0.130	0.006	0.049	0.483	0.013	0.383	0.834	0.908	0.889	0.347	0.010	0.067	0.021	0.332		0.973
ZnNO ₂ Tz	0.180	0.003	0.910	0.603	0.597	0.799	0.416	0.327	0.139	0.006	0.053	0.462	0.014	0.402	0.861	0.881	0.916	0.330	0.011	0.072	0.023	0.350	0.973	

3.2 Time-Resolved or Gated Emission

The spatially averaged, temporally gated emission spectra for each sample from the laser-induced plasma, post-plasma combustion, and self-sustained combustion reactions for each sample are shown in Appendix B, Figures B-1–B-8. The plasma spectra are dominated by the emission from both atomic and ionic species, while the post-plasma spectra are dominated by atomic and some molecular emission—but do not have any significant ionic species emission. The self-sustained combustion spectra primarily exhibit Na emission with strong molecular emission from hydroxyl radical (OH) and manganese^{II} oxide (MnO) (for the Mn12 sample). Typical contaminants include calcium (Ca), lithium (Li), and potassium (K). Minor metal contaminants such as Al, magnesium (Mg), chromium (Cr), or Fe appear in some spectra. All samples have carbon (C), hydrogen (H), nitrogen (N), and oxygen (O) emission in the laser-induced plasma; air entrained into the plasma contributes to the N and O signals. Molecular emission from the cyano radical (CN) persists in both the plasma and post-plasma spectra. Other species from the transition metal components such as Mn, Fe, Co, Ni, Cu, and Zn appear in the corresponding spectra.

3.2.1 Laser-Induced Plasma Emission

The laser-induced plasma emission spectra for all samples with no magnetic field are directly compared in Appendix C, Figure C-1. Figure 14 shows the laser-induced plasma spectra for NaNO₂Tz with and without the magnetic field. While some species such as the Na, Al, and C lines decrease under the magnetic field, others such as H, O, Fe, and Ca increase. The plasma emission intensities for the CuNO₂Tz do not change significantly under the magnetic field, except for an increase in the Fe and Al impurity lines (Figure 15). Emission intensities for most species in the CoNO₂Tz spectra decrease slightly in the presence of the magnetic field, except for the Fe impurity (Figure 16). All emission intensities decrease for FeNO₂Tz in the magnetic field, including the Fe lines (Figure 17). The magnetic field decreases most emission intensities for Mn12—except for the ionic (Mn II) lines (Figure 18). In contrast, the Mn II emission intensities decrease significantly for MnNO₂Tz under the magnetic field (Figure 19). Most emission intensities decrease for NiNO₂Tz except for Ca (Figure 20); both the C and Ca increase for ZnNO₂Tz (Figure 21). The background-corrected peak emission intensities for the organic species are shown in Appendix C, Figure C-2. Only small differences were observed under the magnetic field, with some species' intensities increasing and others decreasing, depending on the sample. Figure 22 shows the total plasma intensities and background-corrected, peak metallic atomic or ionic emission intensities.

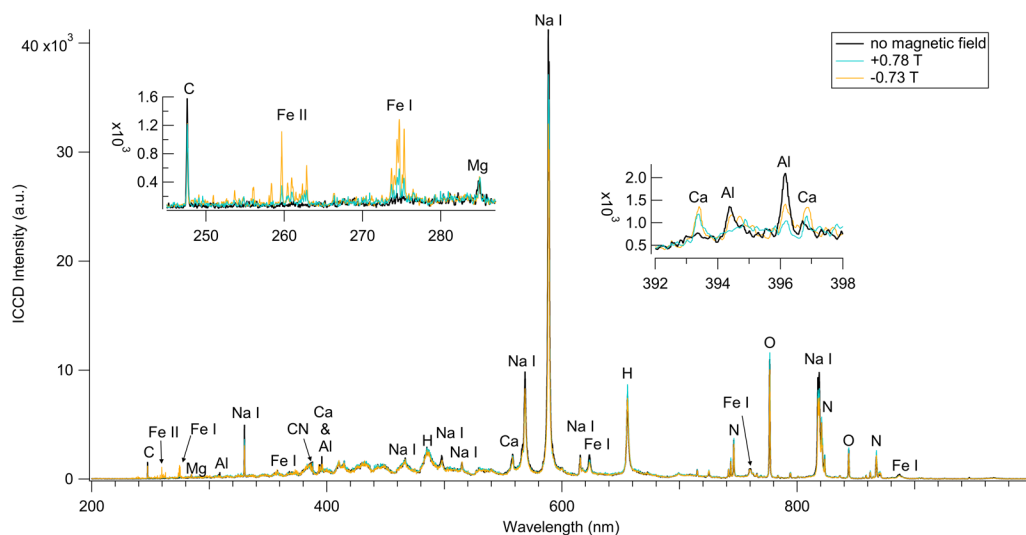


Figure 14. Laser-induced plasma spectra for the NaNO_2Tz with and without an external magnetic field.

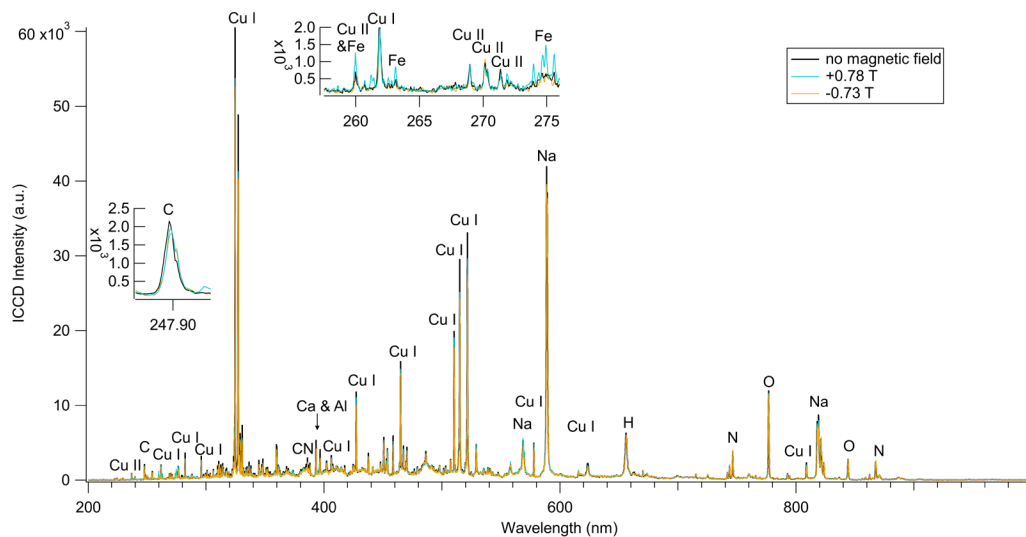


Figure 15. Laser-induced plasma emission spectra for the CuNO_2Tz with and without an external magnetic field.

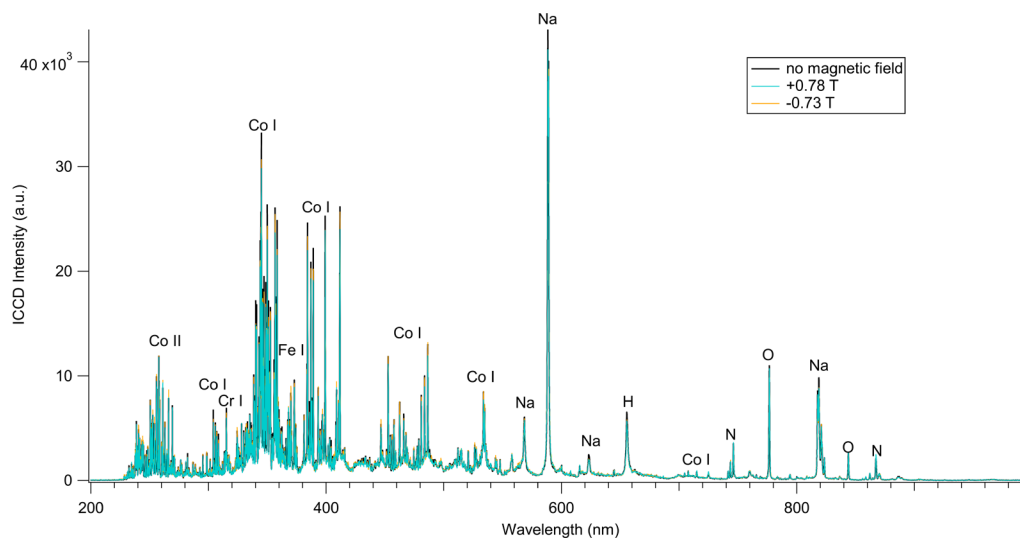


Figure 16. Laser-induced plasma emission spectra for the CoNO_2Tz with and without an external magnetic field.

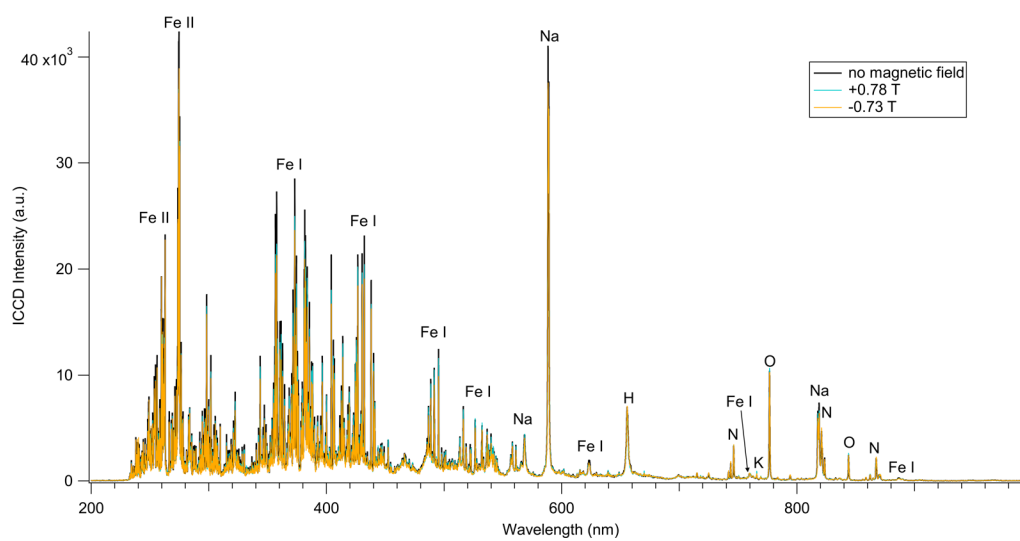


Figure 17. Laser-induced plasma emission spectra for the FeNO_2Tz with and without an external magnetic field.

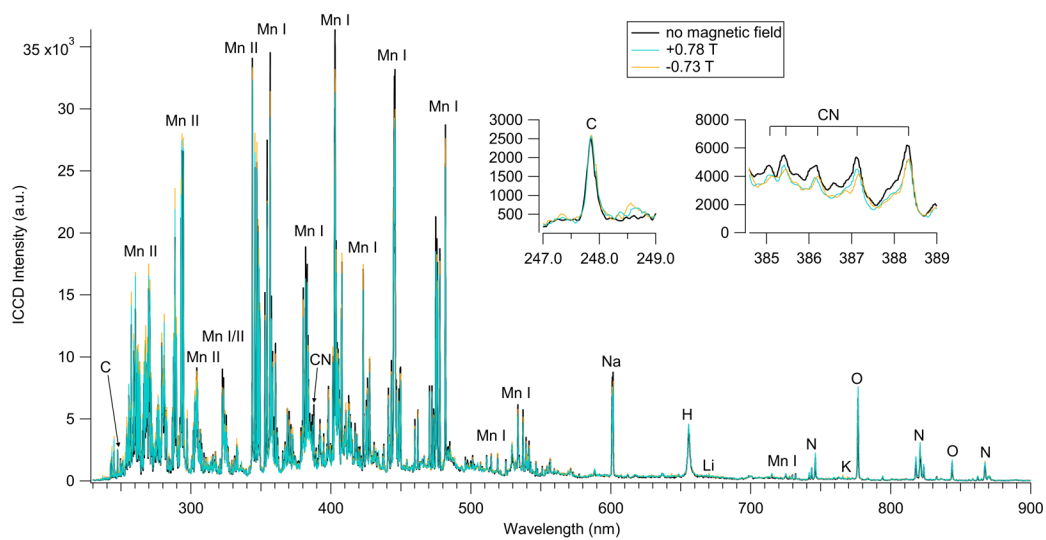


Figure 18. Laser-induced plasma emission spectra for the Mn12 with and without an external magnetic field.

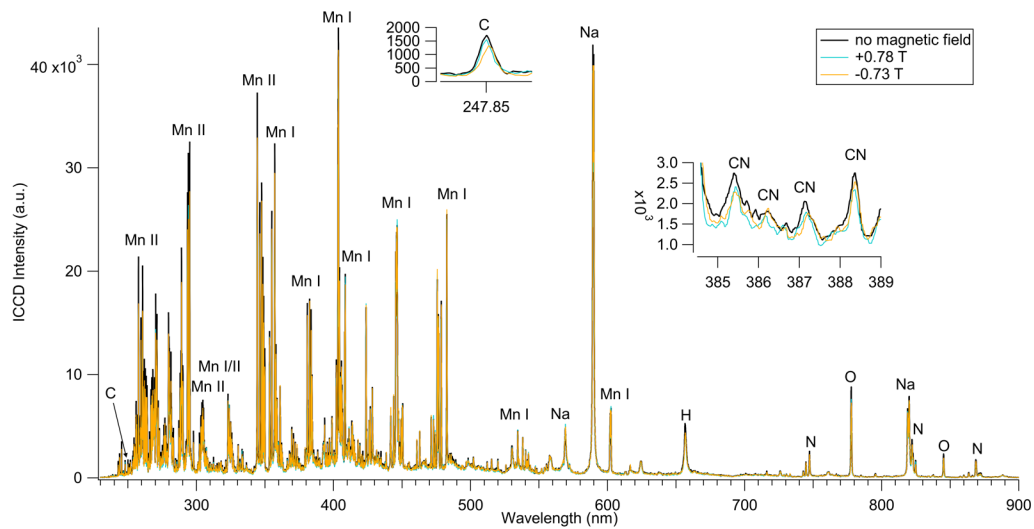


Figure 19. Laser-induced plasma emission spectra for the MnNO₂Tz with and without an external magnetic field.

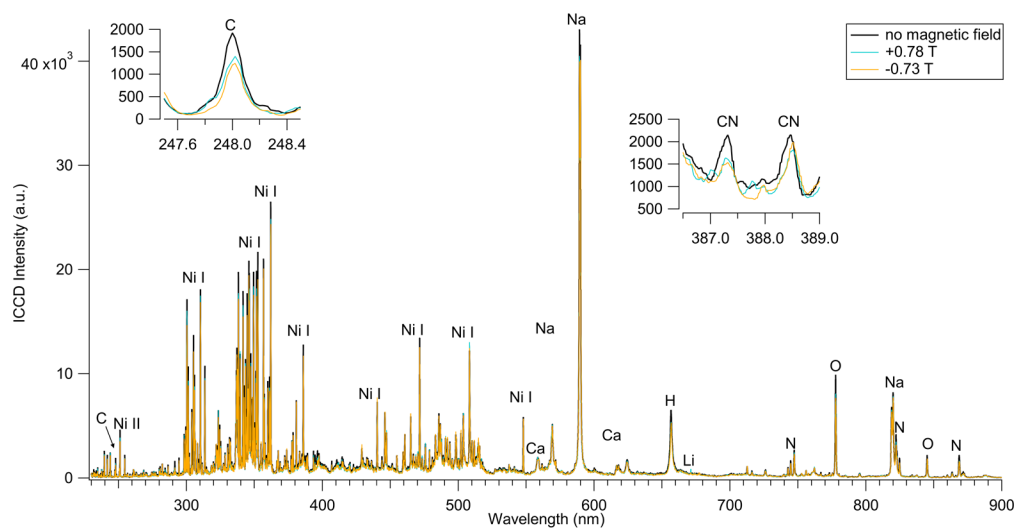


Figure 20. Laser-induced plasma emission spectra for the NiNO_2Tz with and without an external magnetic field.

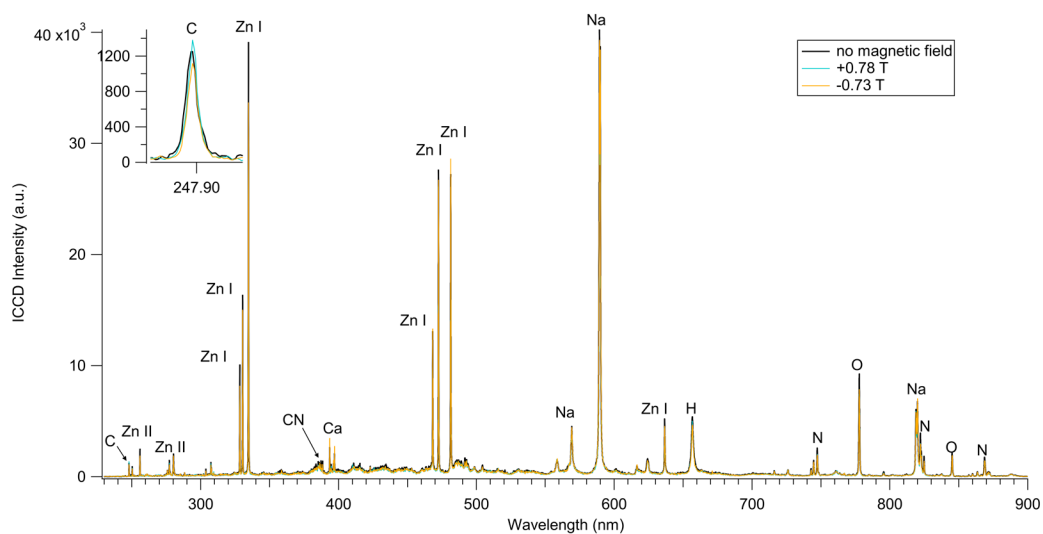


Figure 21. Laser-induced plasma emission spectra for the ZnNO_2Tz with and without an external magnetic field.

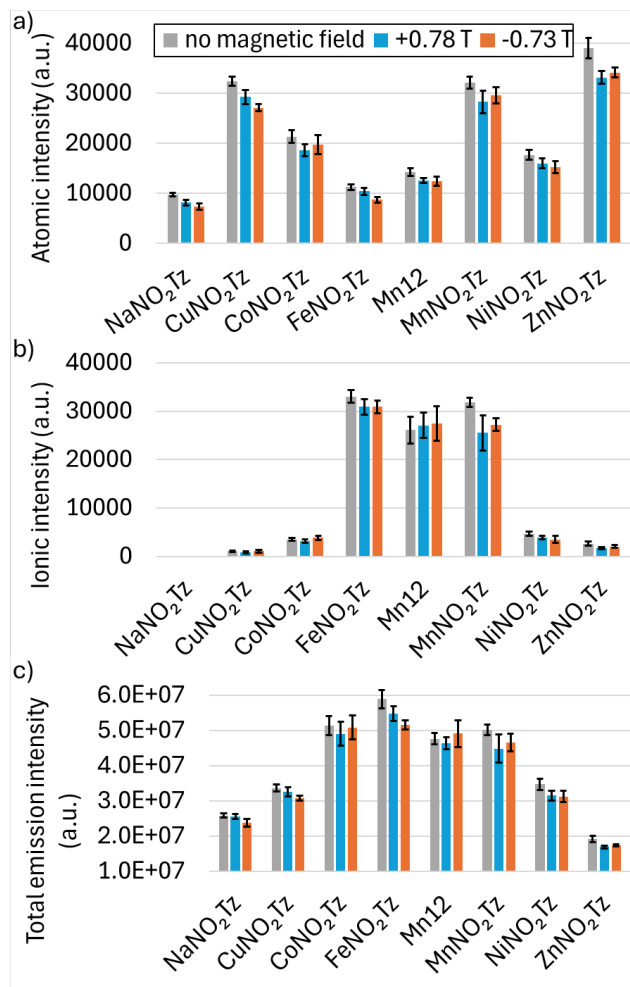


Figure 22. Background-corrected peak a) atomic (neutral), b) ionic (singly ionized), or c) total (summed over all wavelengths) laser-induced plasma emission intensities for each sample with and without an external magnetic field based on the dominant metal component (Na, Cu, Co, Fe, Mn, Ni, or Zn).

The relative emission intensities of the H_α (656 nm), H_β (486 nm), and H_γ (434 nm) lines were used to calculate the plasma temperature using the Boltzmann plot method (Figure 23a). For explosives, these temperatures have been shown to strongly correlate to the detonation temperature.²³ For NaNO_2Tz and MnNO_2Tz , the temperatures consistently decreased under the magnetic field irrespective of field direction; for the other samples, the H temperature sometimes increased or decreased depending on the field direction. The vibrational temperature was also calculated based on the relative intensities of the $\Delta v = 0$ band of the $B^2\Sigma-X^2\Sigma$ violet system of CN (388.22, 387.00, 386.03, 385.32, and 384.91 nm) (Figure 23b). Both NiNO_2Tz and ZnNO_2Tz showed consistent increases in vibrational temperature under the magnetic field, while CuNO_2Tz and Mn12 showed consistent decreases;

interference from metal lines precluded determining the vibrational temperatures for CoNO_2Tz and FeNO_2Tz .

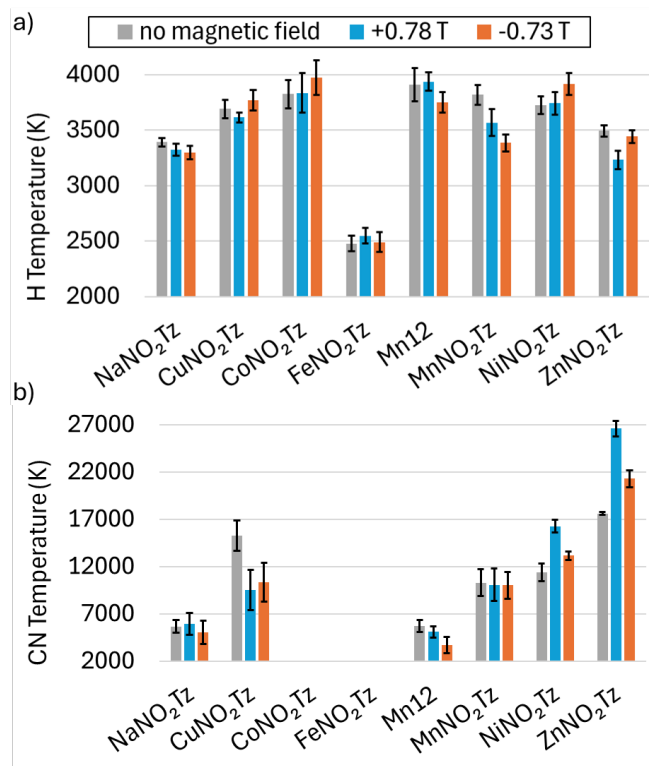


Figure 23. Comparison of a) excitation and b) vibrational plasma temperatures for each sample with and without an external magnetic field calculated based on the relative intensities of the H or CN emission from the laser-induced plasma spectra, respectively.

The dominant metal atomic and ionic emission lines for each sample were used to calculate excitation and ionization temperatures, respectively (Table 4). The results are shown in Figure 24. The metal excitation temperatures increased under the magnetic field for NaNO_2Tz , FeNO_2Tz , MnNO_2Tz , and NiNO_2Tz —but decreased for Mn12 ; the other samples showed no change in temperature. Ionization temperatures decreased consistently for CuNO_2Tz , but either increased or decreased based on field direction for CoNO_2Tz , FeNO_2Tz , MnNO_2Tz , and NiNO_2Tz ; no change in ionization temperature was observed for Mn12 , ZnNO_2Tz , or NaNO_2Tz (for which no Na II lines were present).

Table 4. Metal emission lines used to calculate excitation and ionization temperatures.

Sample	Species	Emission lines for Boltzmann plot
NaNO ₂ Tz	Na I	330.24, 446.48, 498.28, 515.34, 568.82, 616.07 nm
CuNO ₂ Tz	Cu I	282.44, 296.12, 465.11, 521.82, 578.21 nm
	Cu II	236.99, 254.48, 268.93, 336.56 nm
CoNO ₂ Tz	Co I	306.182, 345.35, 356.94, 389.41, 521.27, 536.96, 705.29 nm
	Co II	238.89, 251.12, 263.61, 266.35 nm
FeNO ₂ Tz	Fe I	304.76, 346.59, 404.58 nm
	Fe II	259.94, 275.57 nm
Mn12	Mn I	443.64, 482.35, 525.53 nm
	Mn II	260.57, 270.17, 293.93, 344.20 nm
MnNO ₂ Tz	Mn I	443.64, 482.35, 525.53 nm
	Mn II	260.57, 270.17, 293.93, 344.20 nm
NiNO ₂ Tz	Ni I	385.83, 471.44, 508.05, 547.69 nm
	Ni II	237.54, 243.79, 251.09 nm
ZnNO ₂ Tz	Zn I	307.59, 328.23, 334.50, 636.23 nm
	Zn II	250.20, 255.79, 491.16 nm

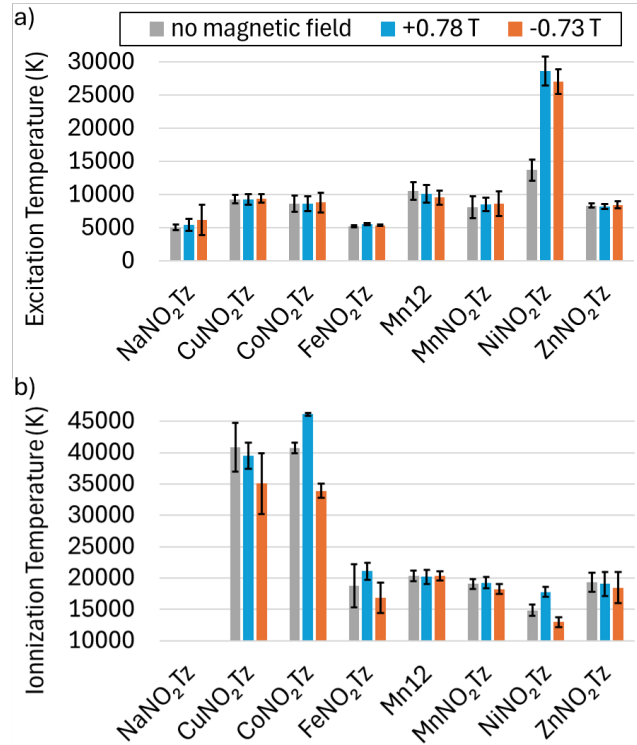


Figure 24. Comparison of a) excitation and b) ionization plasma temperatures for each sample with and without an external magnetic field calculated based on the relative intensities of the dominant metal species emission (i.e., Na, Cu, Co, Fe, or Mn) from the laser-induced plasma spectra, respectively.

As shown in Figure 25a, the ionization ratio (i.e., relative ionic to atomic emission intensity) decreased consistently for NiNO₂Tz and ZnNO₂Tz under the magnetic field and increased consistently for Mn12 and only in the negative field direction

for FeNO₂Tz. In contrast, the electron densities calculated based on the full width at half area of the H_α line using the method of Gigosos et al.²⁵ (Figure 25b) increased for CuNO₂Tz, CoNO₂Tz, MnNO₂Tz, NiNO₂Tz, and ZnNO₂Tz; they decreased consistently for FeNO₂Tz and Mn12, but only in the negative field direction for NaNO₂Tz. This is in contrast to the pure metal powders, for which the electron densities increased regardless of whether the sample was diamagnetic, paramagnetic, or ferromagnetic.¹²

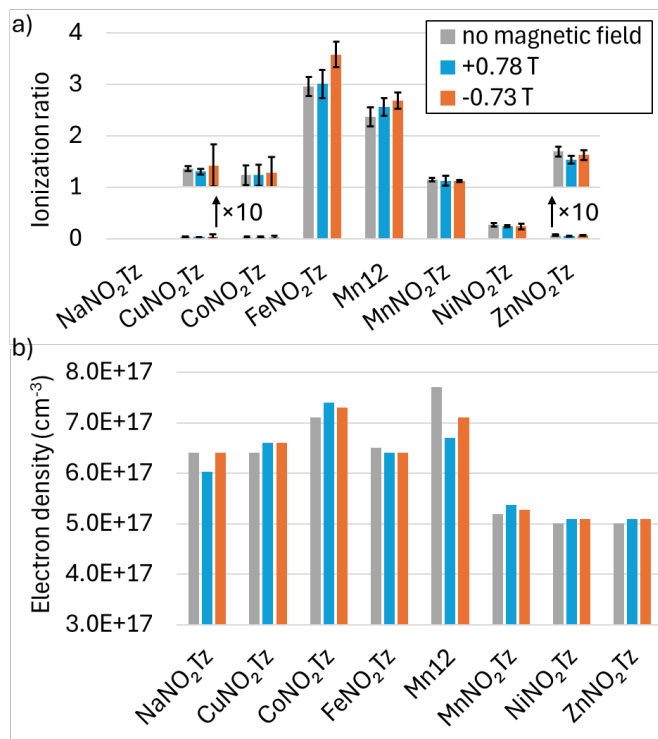


Figure 25. Comparison of the a) ionization ratio based on the dominant metal species (i.e., Na, Cu, Co, Fe, Mn, Ni, or Zn) and b) electron densities calculated based on the H_α line widths from the laser-induced plasma spectra temperatures for each sample with and without an external magnetic field.

Finally, the magnet-induced plasma pressure was calculated based on the red-shift of the Na doublet at 589 nm, which depends on the pressure and temperature.²⁶ In some cases the red-shift had to be estimated because of strong self-absorption of the emission; the Na lines used to determine the red-shift are shown in Appendix C, Figure C-3. The magnet-induced pressures (Figure 26) under both magnetic field directions were similar for all samples except MnNO₂Tz, for which a stronger effect was observed for the lower magnitude, negative field direction.

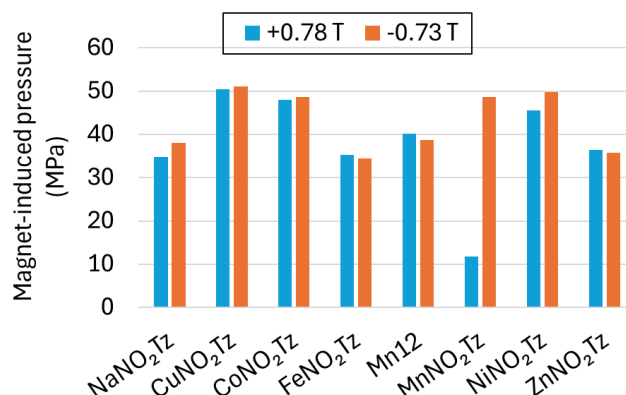


Figure 26. Comparison of the magnet-induced plasma pressure for each sample calculated using the red-shift in the Na line. No red-shift was observed for the samples without the magnetic field.

3.2.2 Post-Plasma Combustion Emission

After approximately 10 μ s, the laser-induced plasma transitions into a cooler, less ionized post-plasma combustion event that can extend for hundreds of microseconds depending on the sample. The reactions in this regime are analogous to those that contribute to the Gurney energy of an explosive.²⁷ The post-plasma combustion emission spectra of all samples (without the magnetic field) are compared in Appendix D, Figure D-1—insets of the Fe and CN lines show interfering species for each sample in the regions of interest. The emission spectra for each individual sample in this regime, with and without the magnetic field, are shown in Figures 27–34. In the post-plasma combustion regime, the emission lines from ionized species are significantly reduced or eliminated, molecular emission generally increases, and emission intensities from impurities such as Na, Ca, K, and Li generally increase as they are continually re-excited during the combustion reactions. As in the laser-induced plasma, emission intensities from metal impurities such as Fe and Al also tend to increase under the magnetic field—although this increase may be dependent on the magnetic field direction. Unlike in the laser-induced plasma regime, CN emission in the post-plasma combustion increases slightly under the magnetic field for several samples. Also, the Fe emission from the FeNO₂Tz sample increases in this regime (Figure 30).

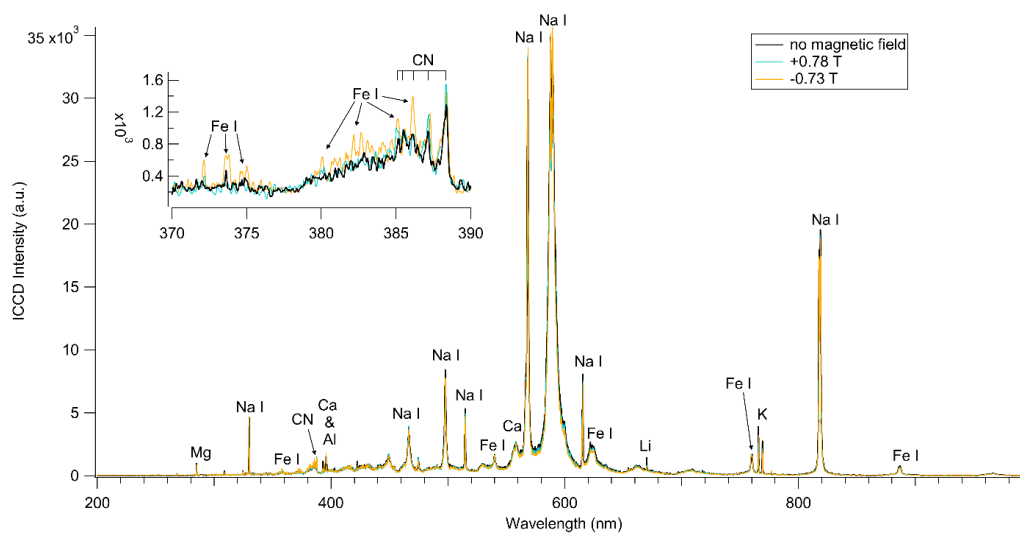


Figure 27. Post-plasma combustion spectra for the NaNO_2Tz with and without an external magnetic field.

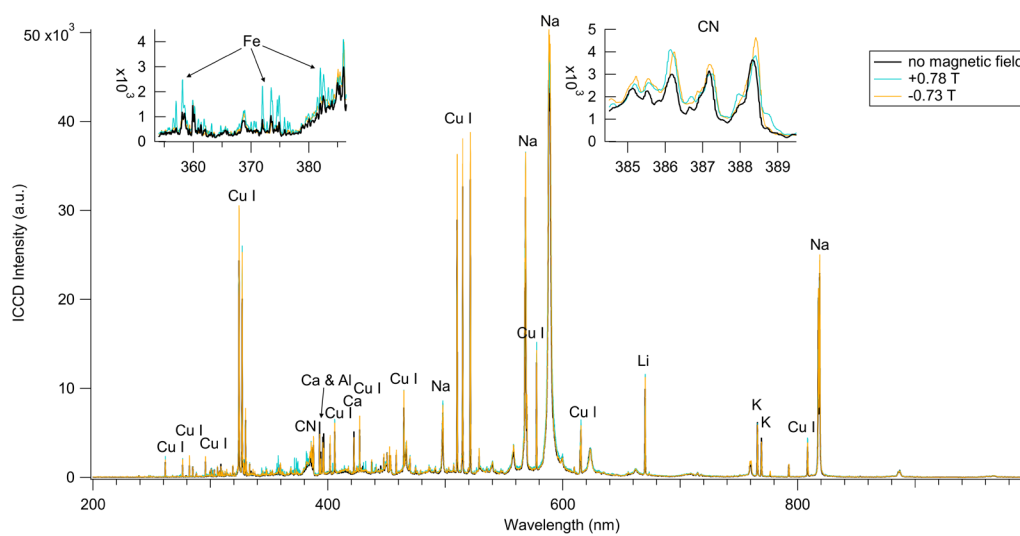


Figure 28. Post-plasma combustion spectra for the CuNO_2Tz with and without an external magnetic field.

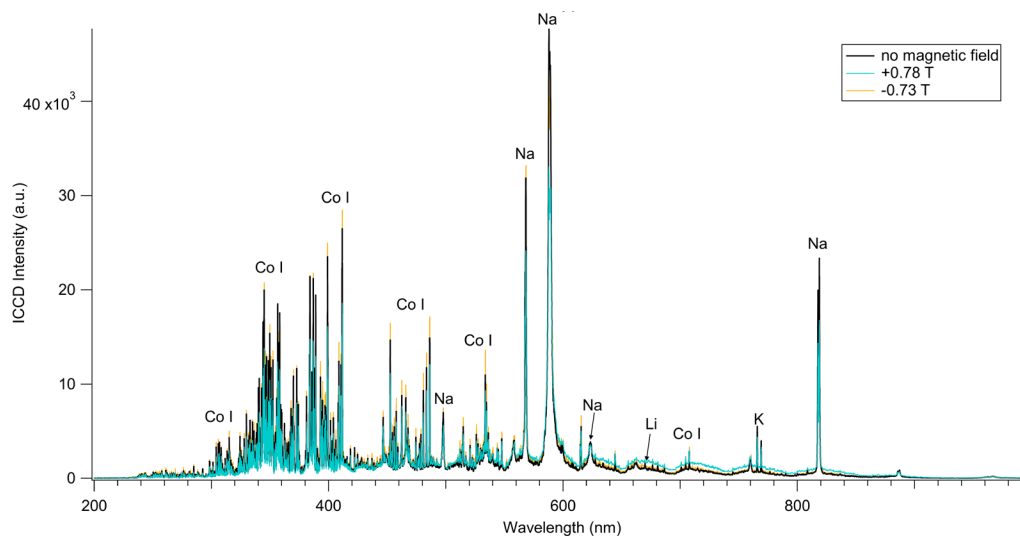


Figure 29. Post-plasma combustion spectra for the CoNO₂Tz with and without an external magnetic field.

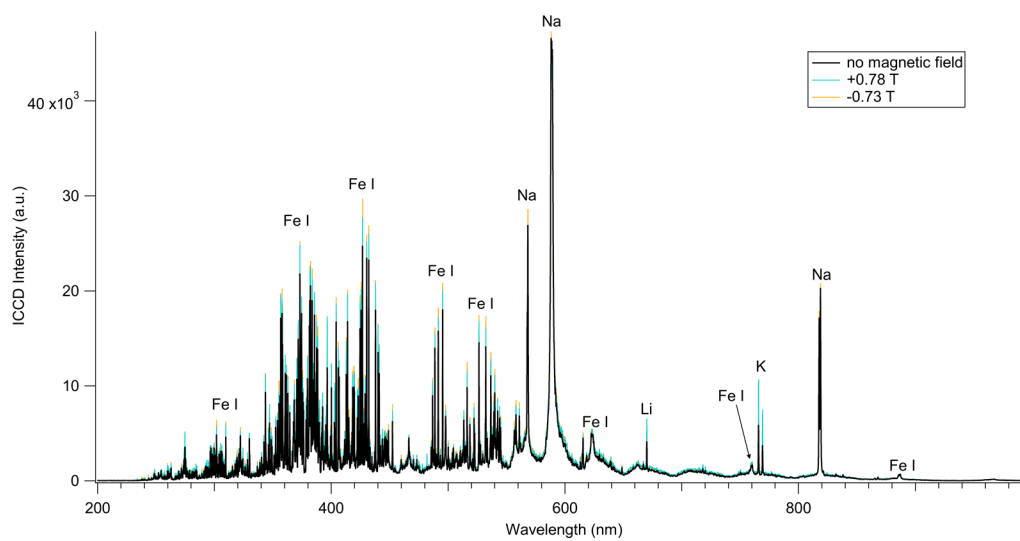


Figure 30. Post-plasma combustion spectra for the FeNO₂Tz with and without an external magnetic field.

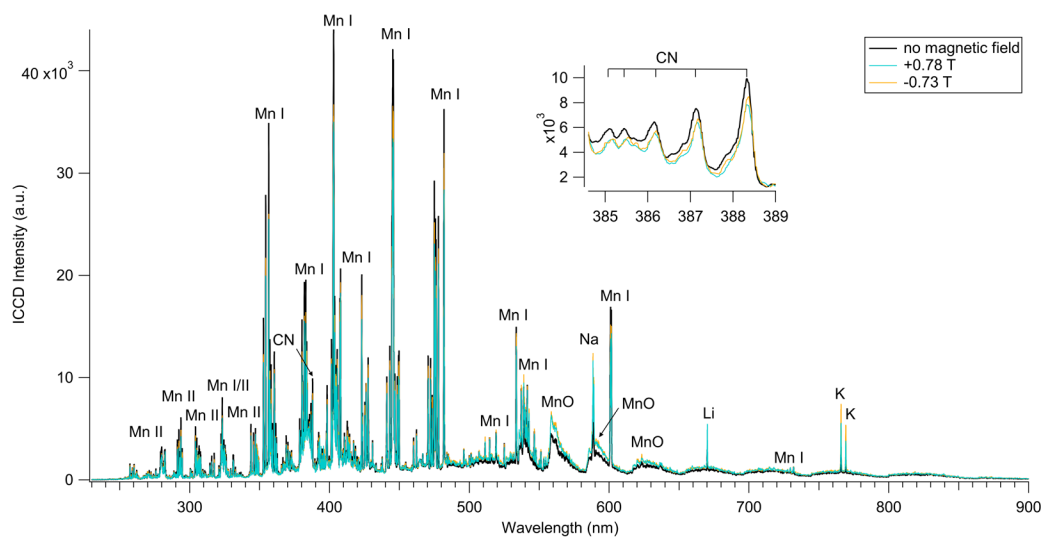


Figure 31. Post-plasma combustion spectra for the Mn12 with and without an external magnetic field.

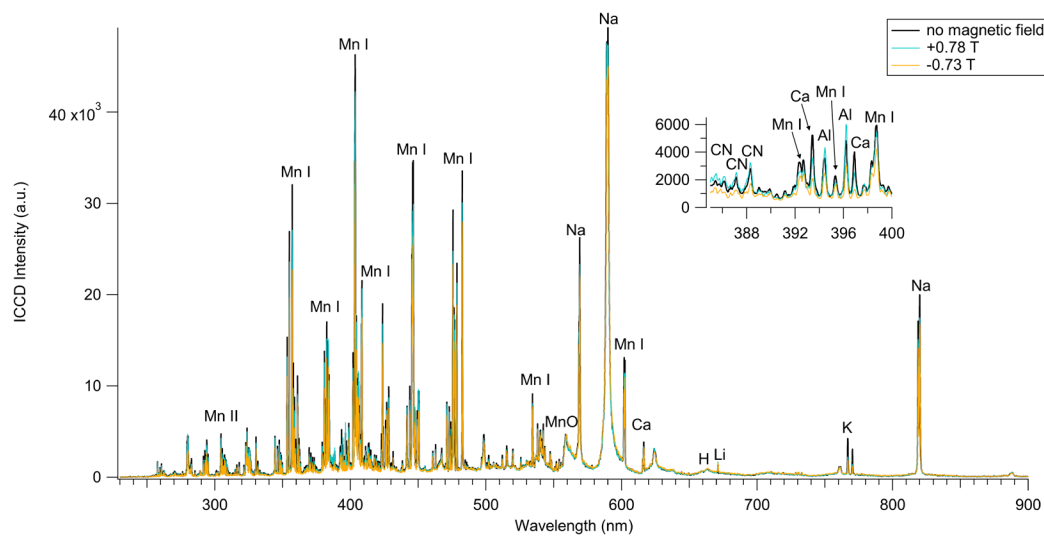


Figure 32. Post-plasma combustion spectra for the MnNO₂Tz with and without an external magnetic field.

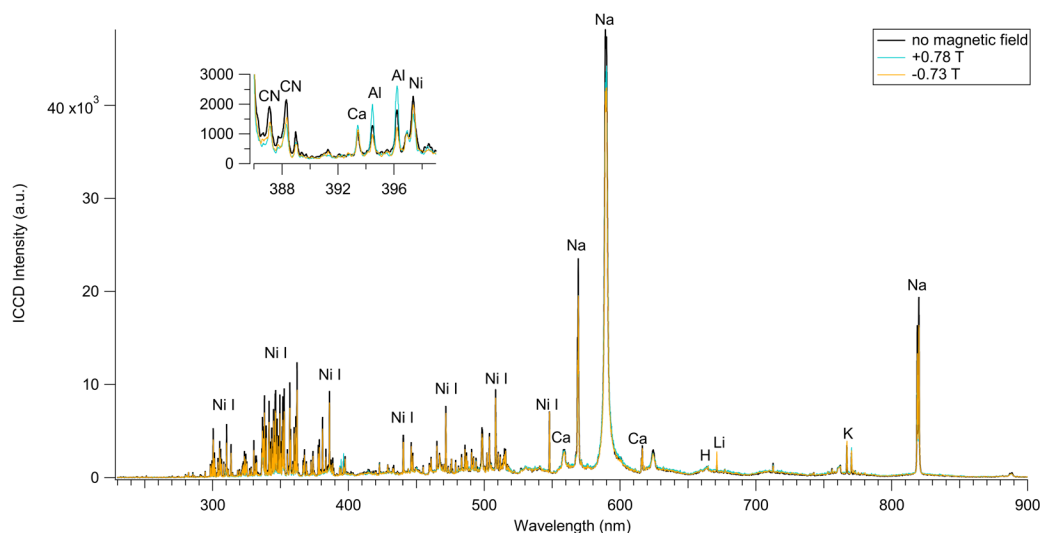


Figure 33. Post-plasma combustion spectra for the NiNO_2Tz with and without an external magnetic field.

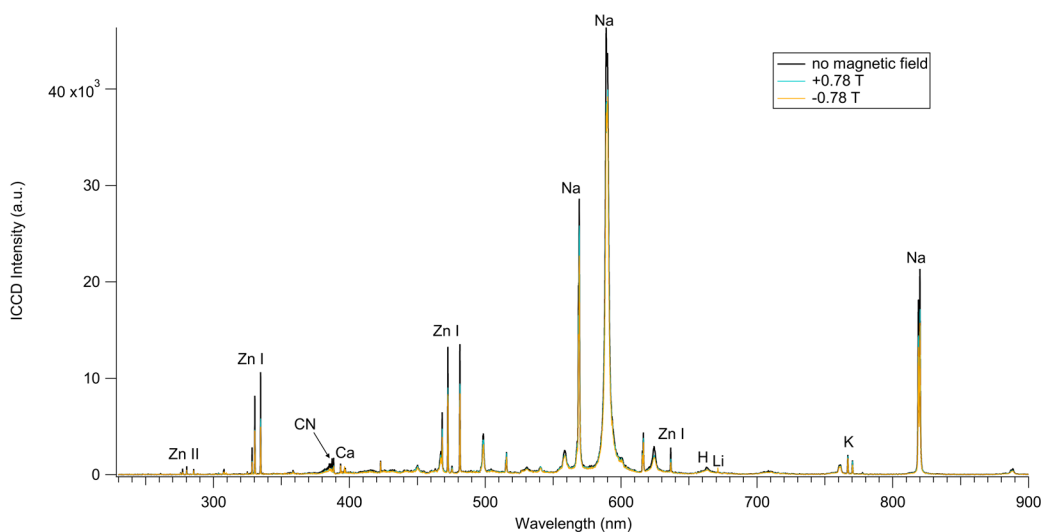


Figure 34. Post-plasma combustion spectra for the ZnNO_2Tz with and without an external magnetic field.

Figure 35 shows the background-corrected, peak emission intensities for CN, MnO, and Na in the post-plasma combustion regime. The average CN intensity increases under the magnetic field (in at least one field direction) for NaNO_2Tz , CuNO_2Tz , FeNO_2Tz , and MnNO_2Tz (Figure 35a). While the MnO intensity decreases slightly for MnNO_2Tz under the magnetic field, it increases significantly for Mn12 in this regime (Figure 35b). Mn12 has the smallest amount of Na, but the emission intensity increases under the magnetic field (Figures 31 and 35c). The Na emission also increases for CuNO_2Tz but generally decreases for the other samples (in contrast to the Ca, Li, and K lines). Figure D-2 compares the atomic and ionic metal

emission intensities for each sample, along with the total emission intensities. The atomic metal emission intensities only increase under the magnetic field in this regime for CuNO_2Tz and FeNO_2Tz , while ionic metal emission intensities only increase consistently in both field directions for FeNO_2Tz . The total post-plasma combustion emission also increases consistently for CuNO_2Tz and FeNO_2Tz , while decreasing consistently for NaNO_2Tz , MnNO_2Tz , NiNO_2Tz , and ZnNO_2Tz .

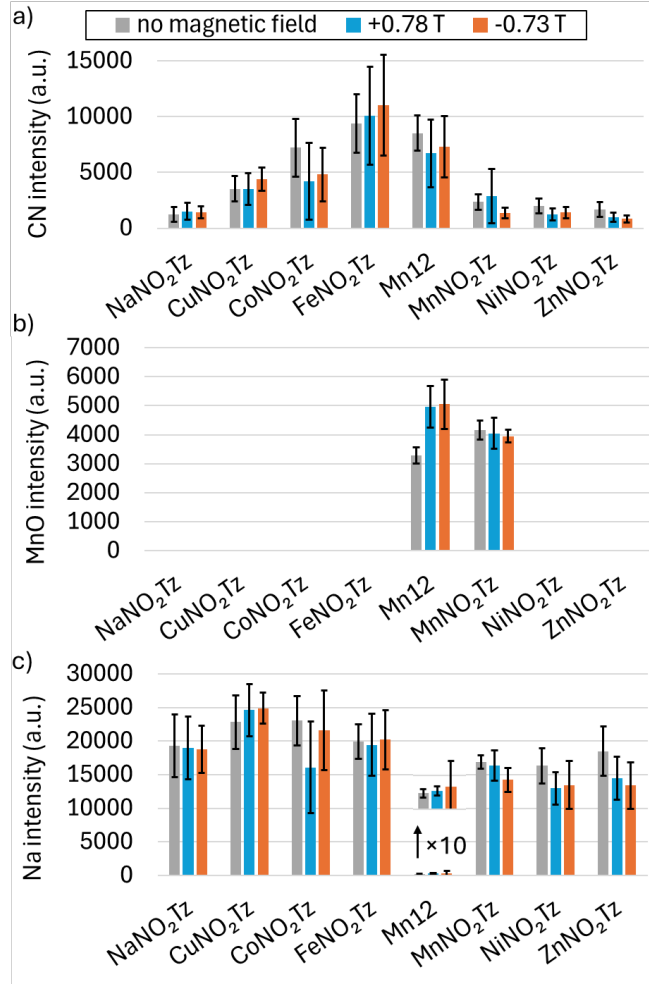


Figure 35. Background-corrected peak emission intensities for a) CN, b) MnO, and c) Na from the post-plasma combustion spectra of each sample with and without an external magnetic field.

The Na lines for each sample in this regime are shown in Appendix D, Figure D-3. The Na doublet near 589 nm showed significant self-absorption in this regime for all samples except Mn12, resulting in self-reversal (i.e., severe absorption of emitted photons by ground-state Na in cooler regions leading to a dip at the line center). In these cases, the nearby Na line at 568.8 nm was used to confirm the red-shift under the magnetic field. The significant self-absorption also reduces the emission intensities of the Na lines, explaining why the Na emission appears to

decrease under the magnetic field (Figure 35c). As shown in Figure 36, the magnet-induced pressure in the post-plasma combustion regime increased for all samples except for CuNO_2Tz (i.e., no magnet-induced shift in the Na lines were observed). The pressures were independent of field direction for all samples except CoNO_2Tz and MnNO_2Tz , for which the negative field direction produced a significantly larger pressure increase.

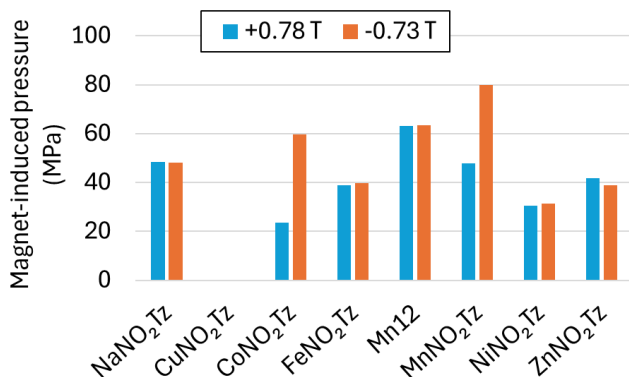


Figure 36. Comparison of the magnet-induced post-plasma combustion pressure for each sample calculated using the red-shift in the Na line. No red-shift was observed for the samples without the magnetic field or for the CuNO_2Tz with the magnetic field.

The time-resolved IR combustion emission traces for each sample with and without the magnetic field were zoomed in to the plasma (saturated signal) and post-plasma combustion region (Figure 37). No significant differences in the post-plasma combustion emission under the magnetic field were observed for NaNO_2Tz , other than a slight delay to peak combustion in the magnetic field direction (Figure 37a). CuNO_2Tz showed no change in post-plasma combustion emission (Figure 37b)—as confirmed by the lack of magnet-induced pressure (see Figure 36). The post-plasma combustion emission increased significantly for CoNO_2Tz (Figure 37c) and ZnNO_2Tz (Figure 37h) under the magnetic field, while it increased slightly at earlier times for FeNO_2Tz (Figure 37d) and Mn12 (Figure 37e). Both MnNO_2Tz (Figure 37f) and NiNO_2Tz (Figure 37g) showed significantly decreased post-plasma combustion in the magnetic field.

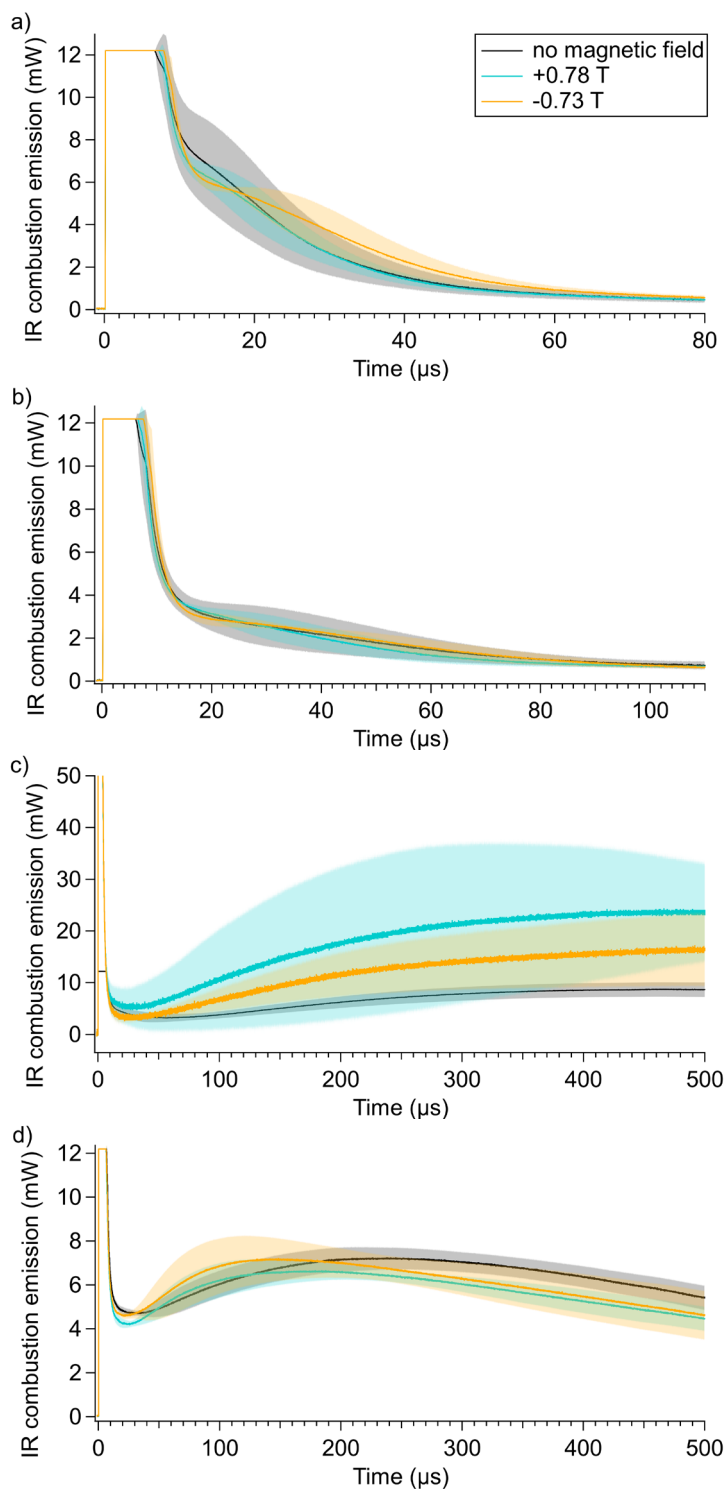


Figure 37. Time-resolved post-plasma combustion emission integrated over the IR region for the a) NaNO₂Tz, b) CuNO₂Tz, c) CoNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz with and without an external magnetic field. Shaded regions are 95% CIs.

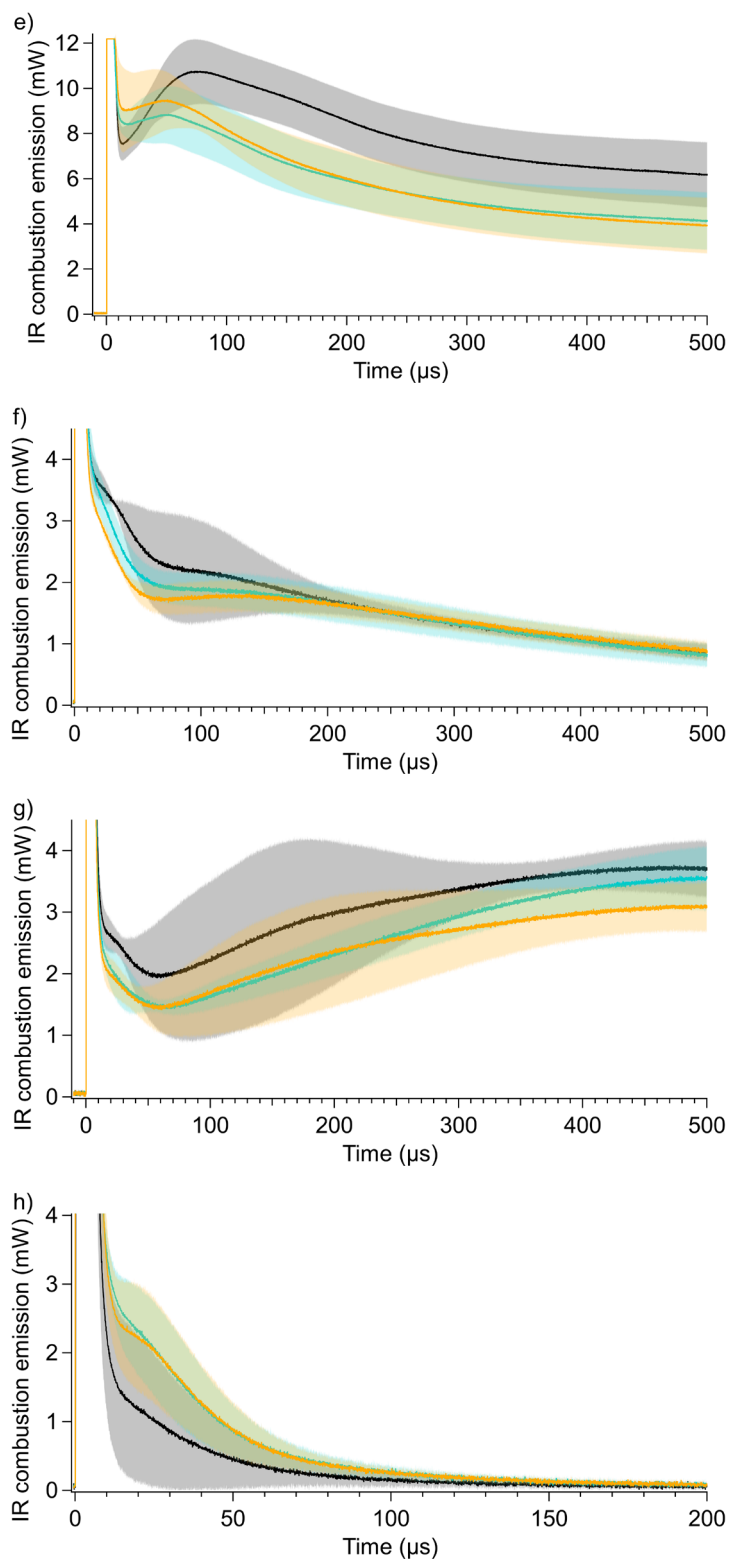


Figure 37. Time-resolved post-plasma combustion emission integrated over the IR region for the a) NaNO_2Tz , b) CuNO_2Tz , c) CoNO_2Tz , d) FeNO_2Tz , e) Mn12 , f) MnNO_2Tz , g) NiNO_2Tz , and h) ZnNO_2Tz with and without an external magnetic field. Shaded regions are 95% CIs (continued).

3.2.3 Self-Sustained Combustion Emission

The self-sustained combustion regime refers to the millisecond-timescale combustion that occurs when the particles ejected from the sample slide reach the air heated by the laser-induced plasma, passage of the laser-induced shock wave, and high-temperature post-plasma combustion reactions and start combusting. Unlike the laser-induced plasma ($\sim 10,000$ K) and post-plasma combustion ($\sim 6,000$ K) regimes, the temperatures in the self-sustained combustion regime are similar to the adiabatic flame temperatures of the materials ($\sim 3,000$ K). This regime correlates to the reactions in a detonation-induced fireball.²⁸ Direct comparisons of the combustion spectra for all samples with or without the magnetic field are shown in Appendix E, Figure E-1. The self-sustained combustion spectra for each sample individually are shown in Figures 38–45. The spectra are dominated by emission from the OH and Na. Only the CoNO₂Tz (Figure 40) and Mn12 (Figure 42) have significant deflagration reactions leading to broadband graybody emission. In this regime, all the emission features are significantly enhanced by the magnetic field. Unlike Mn12, the MnNO₂Tz combustion spectra (Figure 43) have no detectable MnO emission in this time regime.

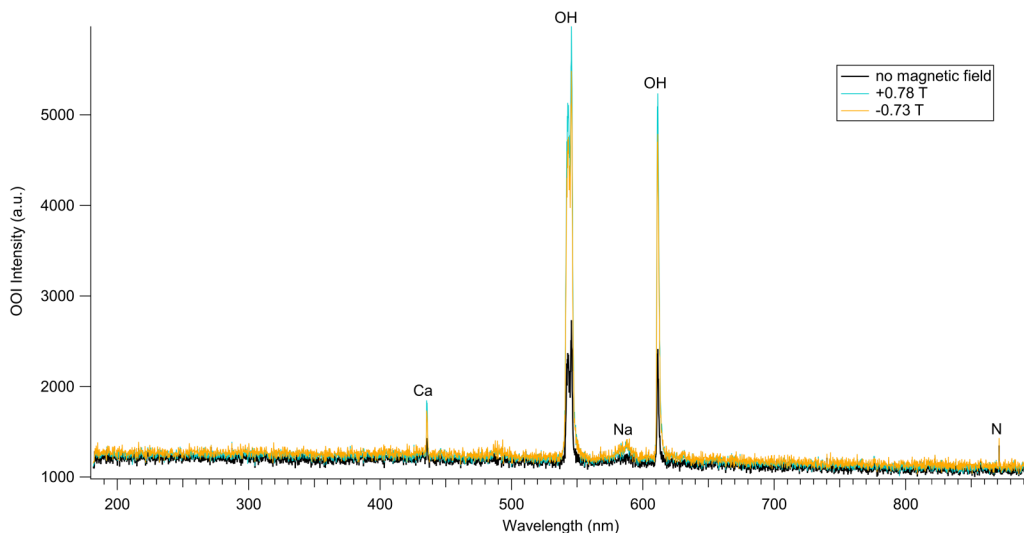


Figure 38. Self-sustained combustion spectra for NaNO₂Tz with and without an external magnetic field.

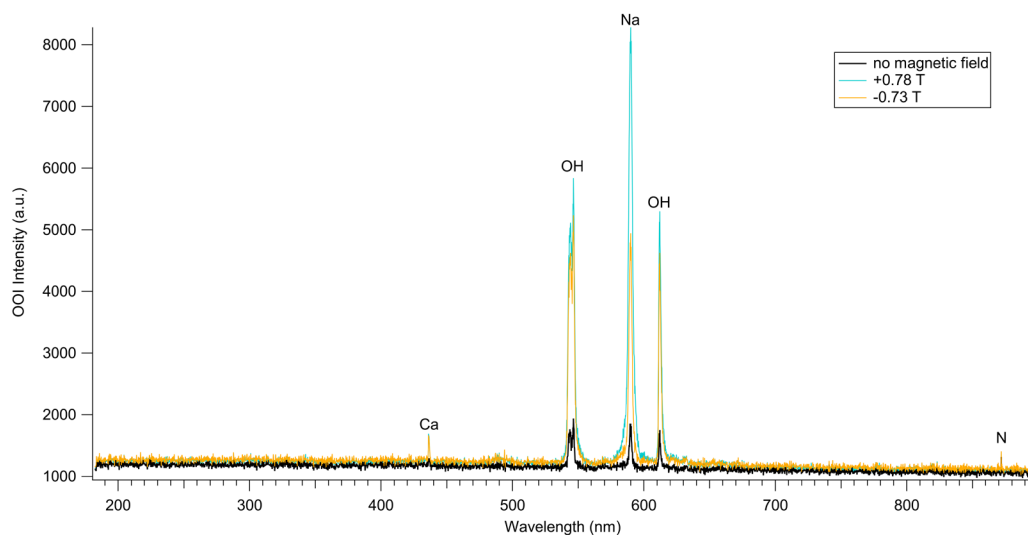


Figure 39. Self-sustained combustion spectra for CuNO_2Tz with and without an external magnetic field.

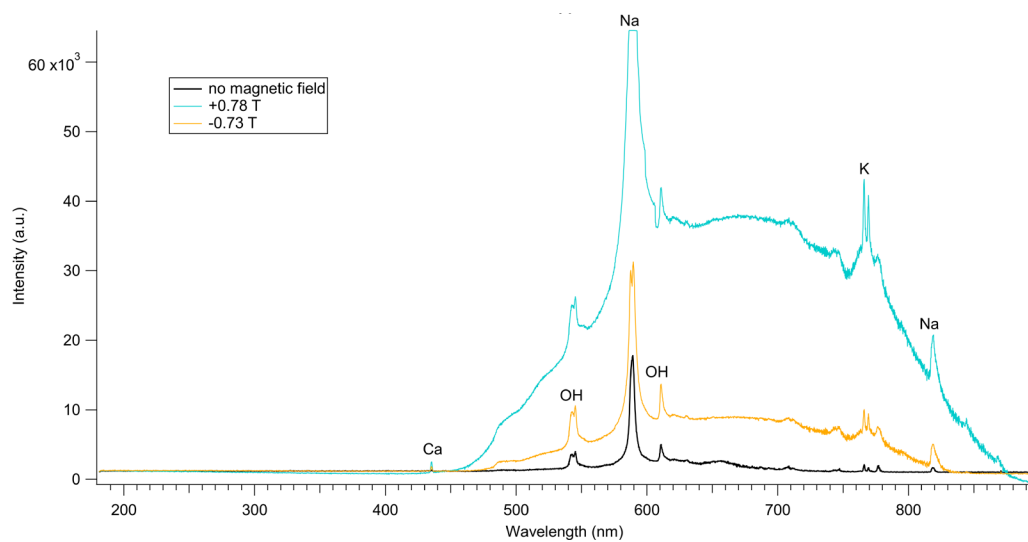


Figure 40. Self-sustained combustion spectra for CoNO_2Tz with and without an external magnetic field.

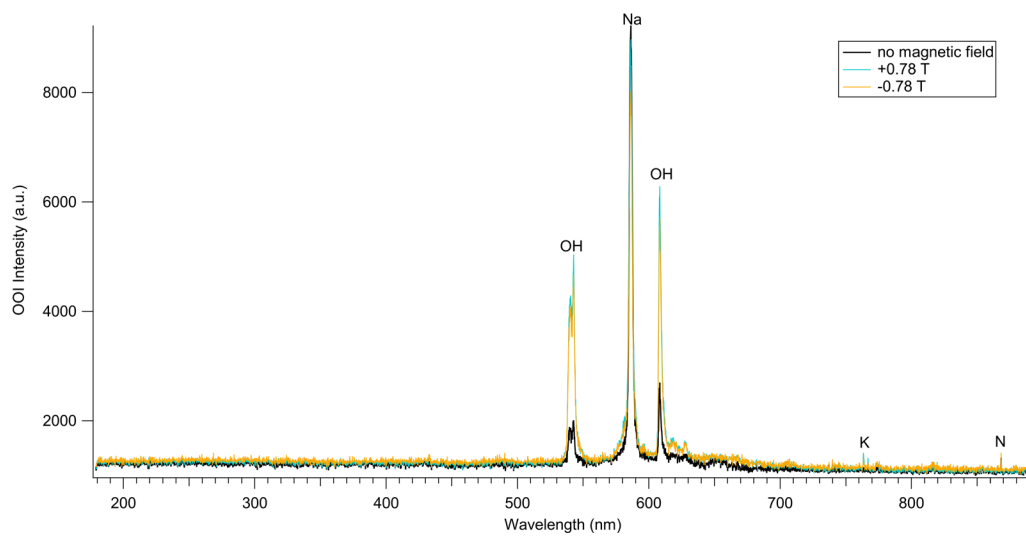


Figure 41. Self-sustained combustion spectra for FeNO_2Tz with and without an external magnetic field.

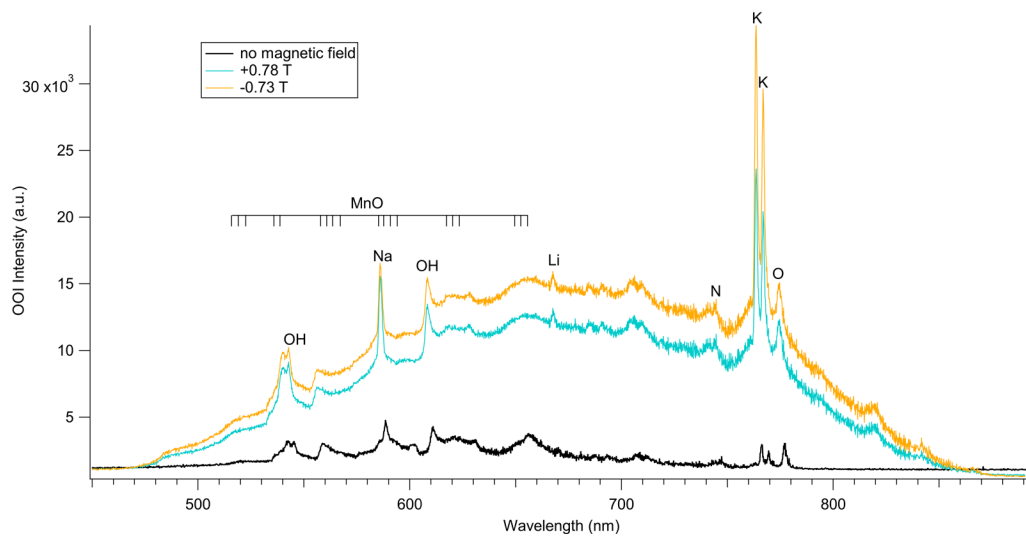


Figure 42. Self-sustained combustion spectra for Mn12 with and without an external magnetic field.

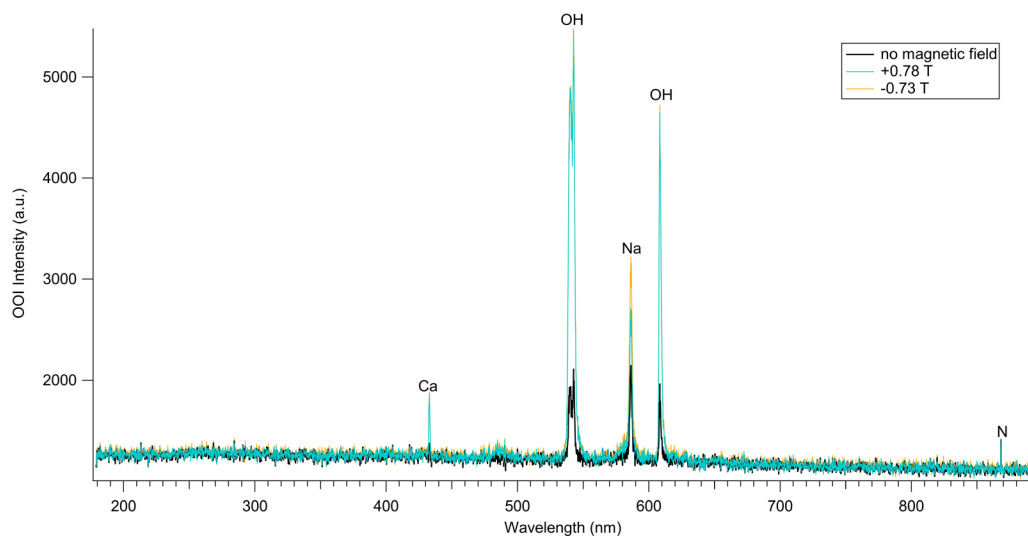


Figure 43. Self-sustained combustion spectra for MnNO_2Tz with and without an external magnetic field.

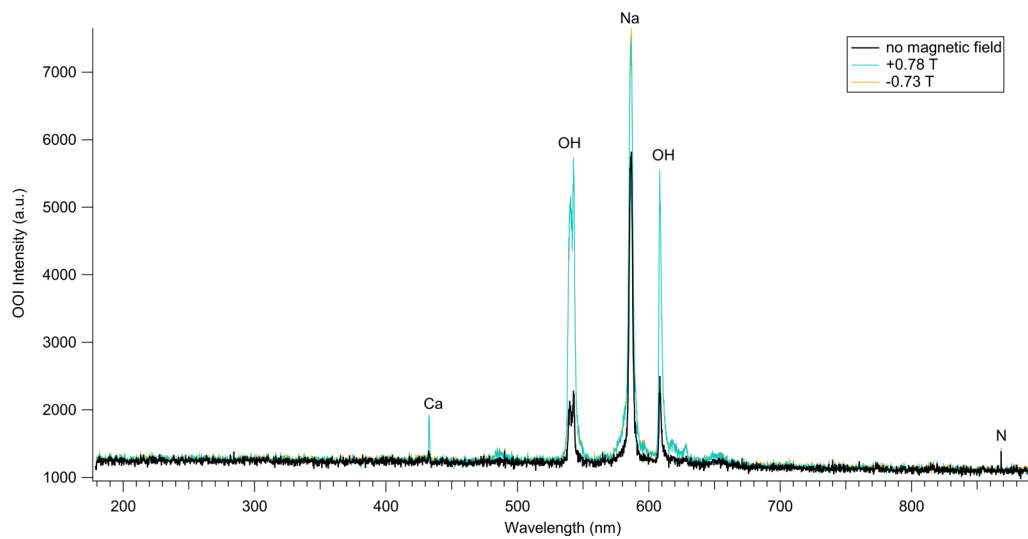


Figure 44. Self-sustained combustion spectra for NiNO_2Tz with and without an external magnetic field.

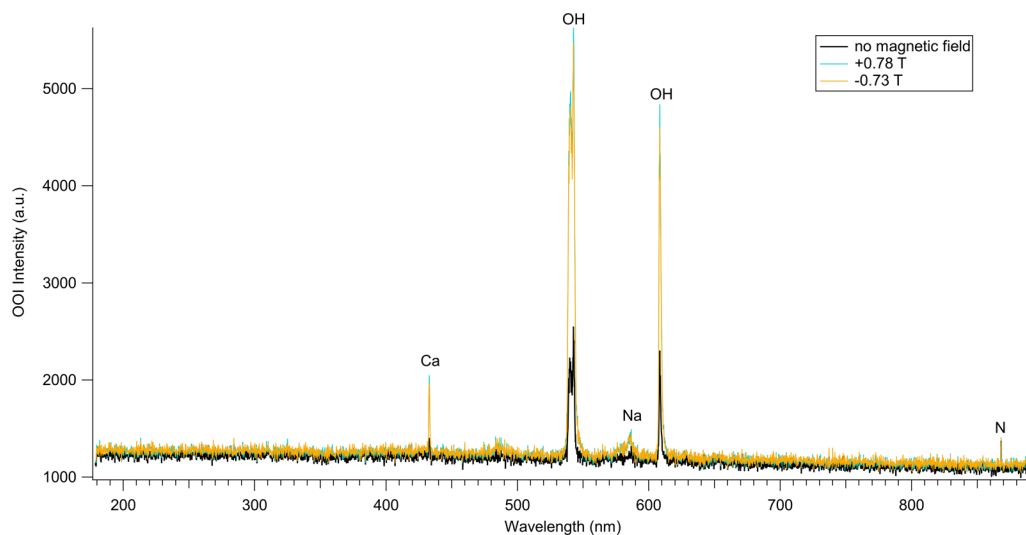


Figure 45. Self-sustained combustion spectra for ZnNO_2Tz with and without an external magnetic field.

The background-corrected, peak emission intensities for the Ca, Li, and K emission are shown in Figure 46. These emission intensities increased (or did not change) for all samples. The Li and K enhancement was particularly strong for CoNO_2Tz and Mn12—while only the positive field direction increased the emission intensity (Ca, Li, K) for CoNO_2Tz .

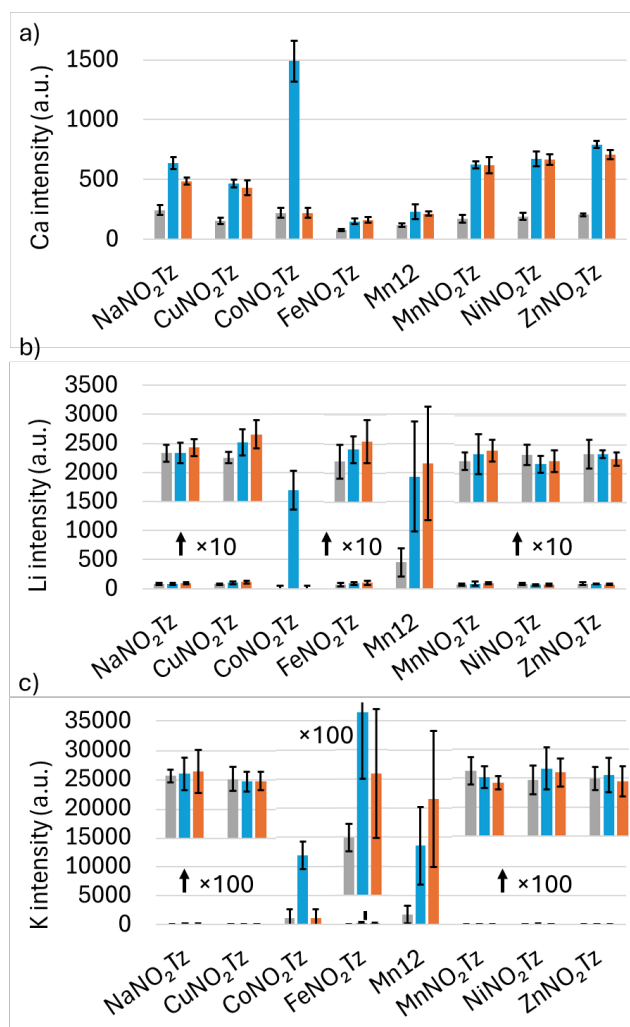


Figure 46. Background-corrected peak emission intensities for a) Ca, b) Li, and c) K from the self-sustained combustion spectra of each sample with and without an external magnetic field.

The emission intensities for the 610-nm OH and 650-nm MnO bands are shown in Figure 47. The OH emission increases for all samples under the magnetic field. The MnO emission only increases for Mn12—no change in peak emission intensity under the magnetic field was observed for MnNO_2Tz . No magnet-induced pressures were calculated because the lack of emission lines (and graybody emission for most samples) precluded temperature calculations; however, no measurable shifts in the Na line position were observed (see Figure E-2).

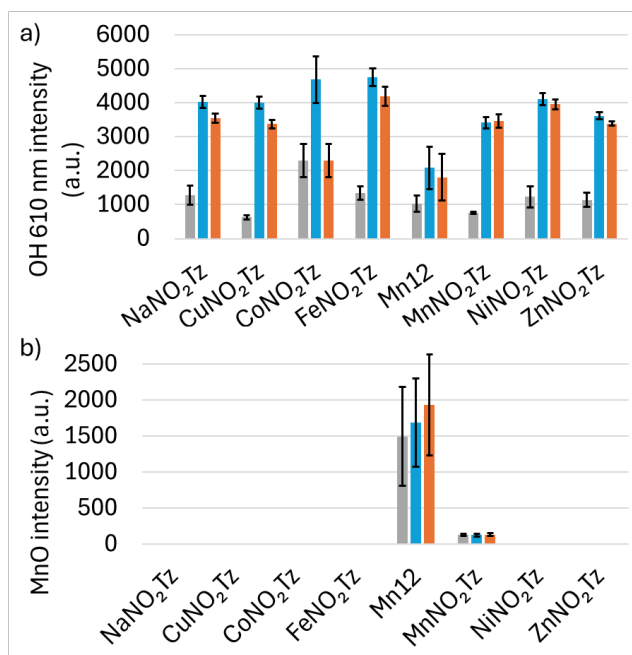


Figure 47. Background-corrected peak emission intensities for a) OH and b) MnO from the self-sustained combustion spectra of each sample with and without an external magnetic field.

The time-resolved IR emission from the combustion reactions on the millisecond timescale are shown in Figure 48. The NaNO₂Tz (Figure 48a), MnNO₂Tz (Figure 48f), and ZnNO₂Tz (Figure 48h) have little or no combustion in this regime (i.e., beyond the post-plasma combustion). The CuNO₂Tz (Figure 48b) shows an increase in self-sustained combustion under the magnetic field, as does the CoNO₂Tz (Figure 48c)—with the biggest increase observed for the CoNO₂Tz. The self-sustained combustion emission decreases for FeNO₂Tz (Figure 48d) and NiNO₂Tz (Figure 48g). The Mn12 combustion emission (Figure 48e) first decreases, then increases after a little over 2 ms. The average IR emission from the self-sustained combustion reactions was integrated over the IR region from 0.8 μ s to 10 ms for each sample and the results are shown in Figure 49. Note that even though the peak NiNO₂Tz combustion emission decreases under the magnetic field, the duration of the combustion is increased and therefore the integrated IR emission does not change significantly.

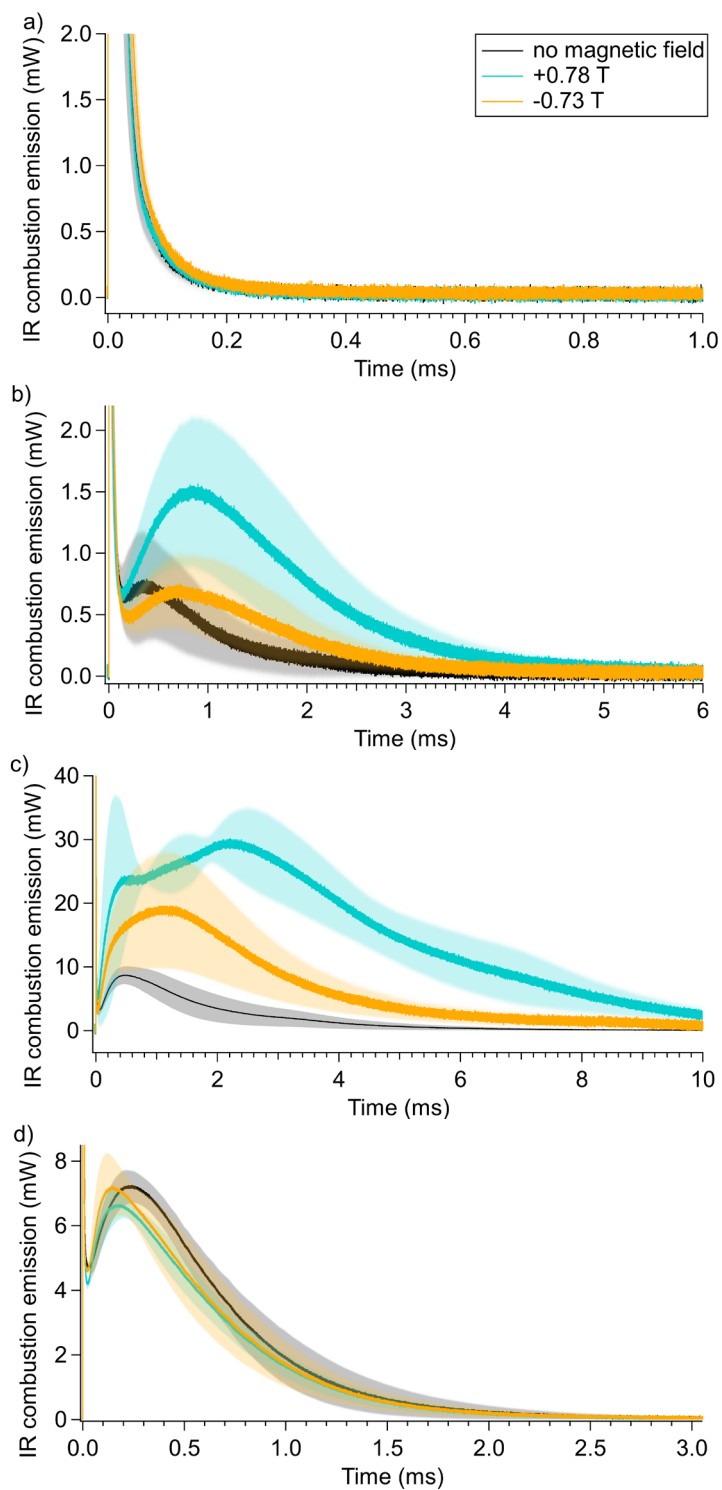


Figure 48. Time-resolved self-sustained combustion emission integrated over the IR region for the a) NaNO₂Tz, b) CuNO₂Tz, c) CoNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz with and without an external magnetic field. Shaded regions are 95% CIs.

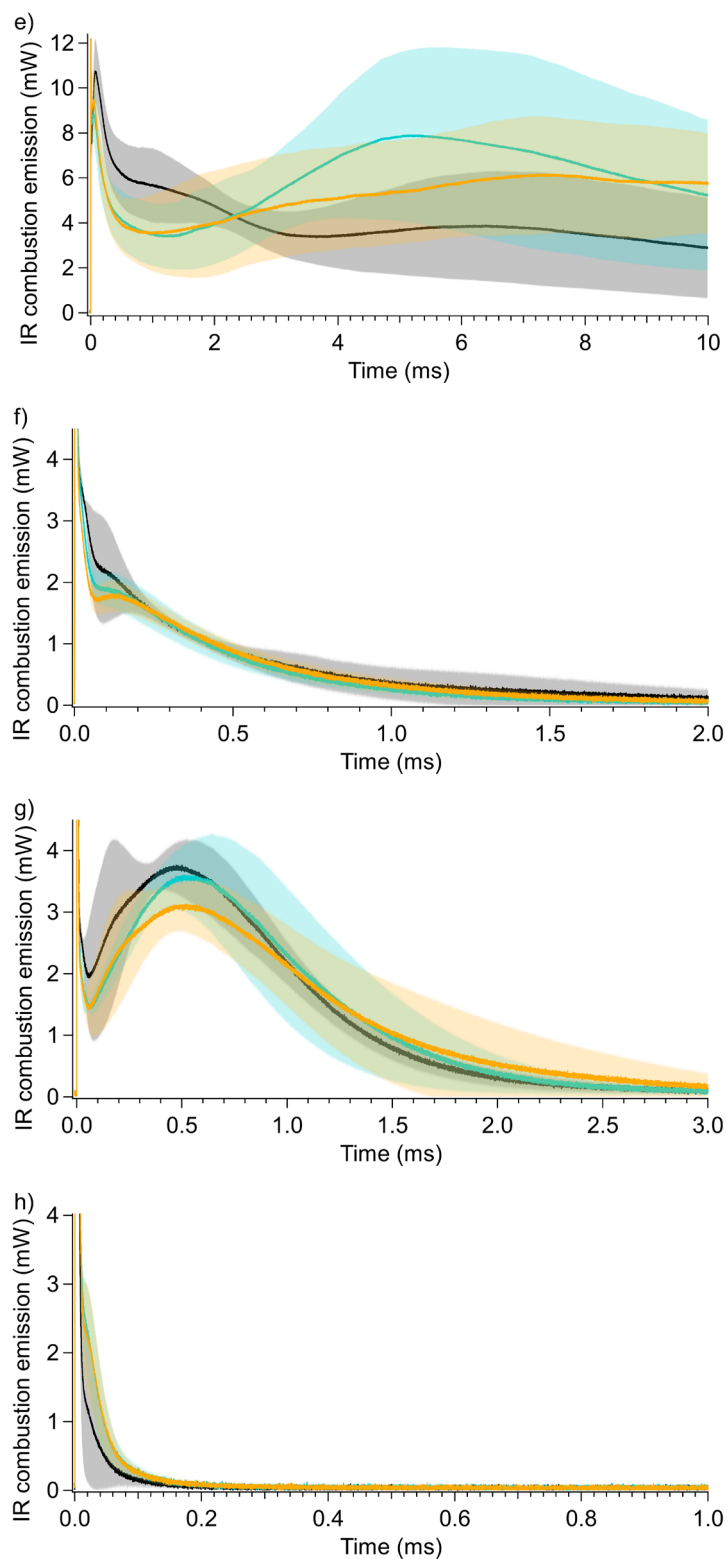


Figure 48. Time-resolved self-sustained combustion emission integrated over the IR region for the a) NaNO₂Tz, b) CuNO₂Tz, c) CoNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz with and without an external magnetic field. Shaded regions are 95% Cis (continued).

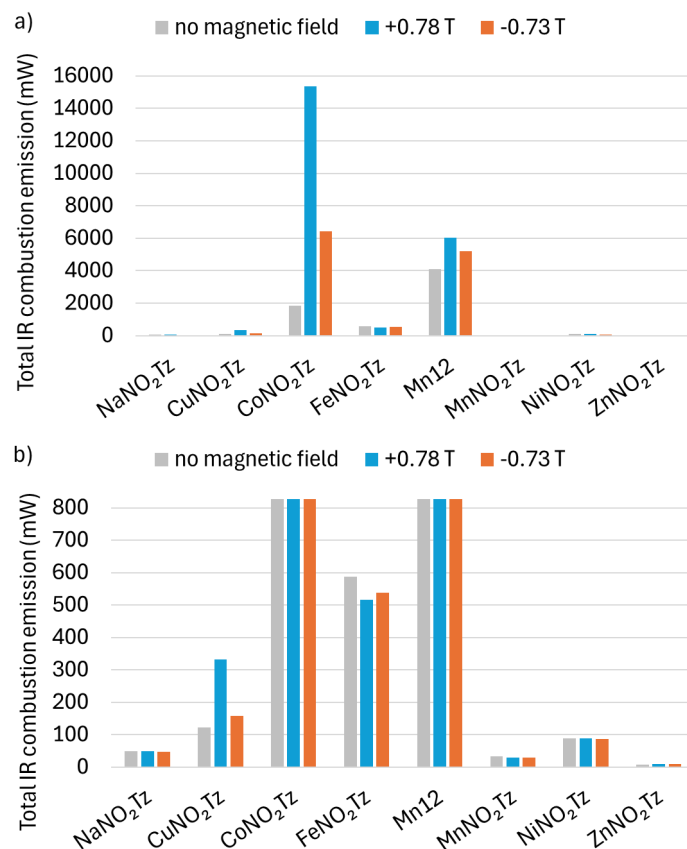


Figure 49. Total IR emission from the self-sustained combustion integrated over the IR region from 0.8 μ s to 10 ms at a) full-scale and b) zoomed-in intensity scale ($\times 20$).

4. Discussion

Table 5 summarizes how the magnetic field influences each measured parameter in the high-speed schlieren videos or during the laser-induced plasma, post-plasma combustion, or self-sustained combustion regimes. Other than the increased total visible emission in the schlieren images, the increased magnet-induced pressure in the laser-induced plasma, the increased overall post-plasma emission intensities, and the decreased metal atom emission in the laser-induced plasma, no trends were consistent among all sample types. Unlike for the neat Fe powders,¹² the Fe emission intensities decreased in the FeNO₂Tz sample under the magnetic field. For most samples and parameters, the trends were independent of field direction—although there were a few exceptions, as noted in the results section. Some of the apparent field direction effects may be a result of small changes and large shot-to-shot variations; however, several of the noted effects were quite large (e.g., the significantly higher increase in magnet-induced pressure for some samples in the -0.73 -T field compared with the $+0.78$ -T field).

Table 5. Summary of the influence of the magnetic field on each sample for each measured parameter.

Parameter	NaNO ₂ Tz		CuNO ₂ Tz		CoNO ₂ Tz		FeNO ₂ Tz		Mn12		MnNO ₂ Tz		NiNO ₂ Tz		ZnNO ₂ Tz	
	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T	+0.78 T	-0.73 T
Characteristic laser-induced shock velocity	↓	↓	↓	↓	↑	↑	↓	↓	↓	↓	↑	↑	↑	↑	↓	↓
Dewey shock velocity	↑	↑	---	---	↑	↑	↓	↓	↑	↑	↑	↑	↓	↓	---	---
Plume heights	↓	↓	↓	↓	↑	---	↓	↓	↓	↓	↑	↑	↓	↓	---	---
Total emission (video)	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑
Total plasma emission (spectra)	↓	↓	↓	↓	↓	↓	↓	↓	↓	↑	↓	↓	↓	↓	↓	↓
Metal atom emission	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓
Metal ion emission	n/a	n/a	↓	---	↓	↑	↓	↓	↑	↑	↓	↓	↓	↓	↓	↓
CN emission	---	↓	↓	↓	---	↓	---	↓	↓	↓	↓	↓	↓	↓	↓	↓
Fe impurity emission	↑	↑	↑	---	---	↑	↓	↓	---	↑	---	↑	---	↑	n/a	n/a
H temperature	↓	↓	↓	↑	---	↑	↑	---	↑	↓	↓	↓	---	↑	↓	↓
CN temperature	↑	↓	↓	↓	n/a	n/a	n/a	n/a	↓	↓	---	---	↑	↑	↑	↑
Metal excitation temperature	↑	↑	---	---	---	---	↑	↑	↓	↓	↑	↑	↑	↑	---	---
Metal ionization temperature	n/a	n/a	↓	↓	↑	↓	↑	↓	---	---	↑	↓	↑	↓	---	---
Ionization ratio	n/a	n/a	---	---	---	---	---	↑	↑	↑	---	---	↓	↓	↓	↓
Electron density	---	↓	↑	↑	↑	↑	↓	↓	↓	↓	↑	↑	↑	↑	↑	↑
Magnet-induced pressure	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑
Total post-plasma combustion (spectra)	↓	↓	↑	↑	---	↑	↑	↑	---	↑	↓	↓	↓	↓	↓	↓
Metal atom emission	---	---	↑	↑	↓	---	↑	↑	↓	↓	↓	↓	↓	↓	↓	↓
Metal ion emission	n/a	n/a	↓	↑	↓	↑	↑	↑	↓	↓	↑	↓	↓	---	↓	↓
CN emission	↑	↑	---	↑	↓	↓	↑	↑	↓	↓	↑	↓	↓	↓	↓	↓
MnO emission	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	↑	↑	---	↓	n/a	n/a	n/a	n/a
Fe impurity emission	↑	↑	↑	↑	n/a	n/a	↑	↑	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
Al impurity emission	↓	↑	↓	↓	n/a	n/a	↑	↑	↑	↑	↑	↓	↑	↓	↓	↑
Magnet-induced pressure	↑	↑	---	---	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑
IR emission	---	↑	---	---	↑	↑	↑	↑	↑↓	↑↓	↓	↓	↓	↓	↑	↑
Total self-sustained combustion (spectra)	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑
OH emission	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑
MnO emission	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	↑	↑	---	---	n/a	n/a	n/a	n/a
IR emission	---	---	↑	↑	↑	↑	↓	↓	↓↑	↓↑	---	---	↓	↓	---	---

Only the CoNO₂Tz and MnNO₂Tz show an increase in microsecond-timescale energy release throughout the plasma lifetime under the magnetic field, although this effect is not very large. NiNO₂Tz shows an increase in very early energy release, while NaNO₂Tz and Mn12 show later increases in microscale energy release—likely from delayed combustion reactions. While the overall plasma emission intensities decrease under the magnetic field, emission lines from certain impurity species (e.g., Al and Fe) are enhanced. Unlike with the neat metal powders,¹² the temperatures and electron densities do not uniformly increase under the magnetic field but depend on the sample. Magnet-induced pressure increases were observed for all samples in the laser-induced plasma and post-plasma combustion regimes. Post-plasma combustion enhancement by the magnetic field was observed for all samples except MnNO₂Tz and NiNO₂Tz. Significant self-sustained combustion enhancement was observed for CoNO₂Tz and Mn12 (and CuNO₂Tz to a lesser extent).

Explanations for the observed trends require detailed characterization of the transition metal energetic complexes—including their magnetic properties—that are not yet available. However, the LASEM results pose several interesting questions. For example, why do Mn12 and MnNO₂Tz show very different energy release behavior under the magnetic field? The plume heights increase for MnNO₂Tz but decrease for Mn12. The ionic emission in the plasma increases for Mn12 but decreases for MnNO₂Tz in the magnetic field—Mn12 also has a significant decrease in electron density in the magnetic field, while the electron density increases for MnNO₂Tz. The Mn12 produces more MnO (and overall combustion) in the magnetic field than MnNO₂Tz—but the enhancement of the combustion emission for Mn12 is modulated (i.e., alternately increases and decreases under the magnetic field) throughout the entire reaction process. These differences are likely related to the decomposition mechanisms for the samples. Other questions include why the emission intensities for Fe impurities increase significantly in the magnetic field, while the Fe emission intensities from FeNO₂Tz decrease. Also, vibrational (CN) temperatures increase significantly only for NiNO₂Tz and ZnNO₂Tz under the magnetic field—which could indicate differences in reaction mechanisms compared with the decomposition of the other NO₂Tz samples.

5. Conclusions

The influence of the magnetic field on the microsecond-timescale energy release was relatively small for all samples—but much more significant effects were observed for the combustion reactions, particularly on the millisecond timescale. Overall, the results demonstrate that a magnetic field can be used to influence the rate of energy release from transition metal energetic complexes. However, the interaction of the magnetic field with the high-temperature reactions of the materials is still poorly understood and requires more detailed characterization of the sample properties and accurate chemical kinetic models (which do not yet exist). Incorporation of these materials into energetic formulations containing explosives would likely alter their responses to the magnetic field.

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Appendix A. Snapshots from the High-Speed Schlieren Videos

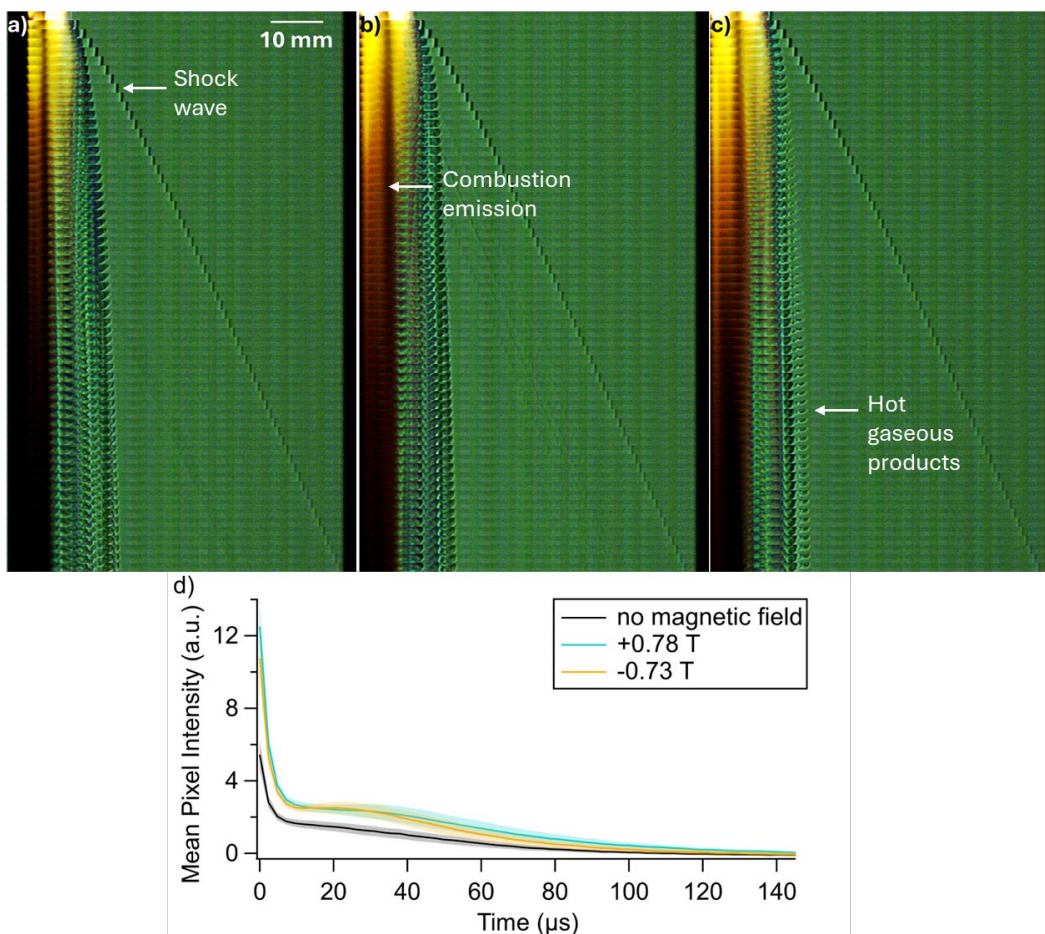


Figure A-1. Snapshots from representative high-speed schlieren videos of the a) NaNO₂Tz, b) NaNO₂Tz (+0.78 T), and c) NaNO₂Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

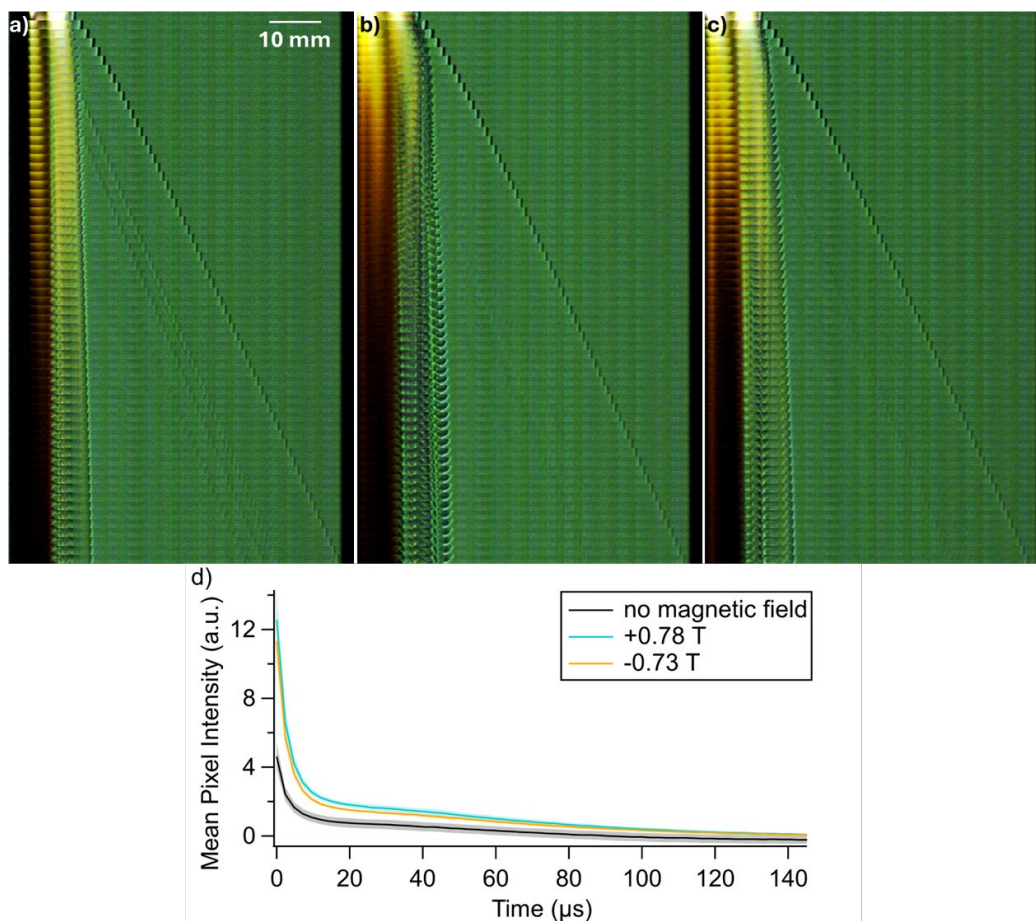


Figure A-2. Snapshots from representative high-speed schlieren videos of the a) CuNO₂Tz, b) CuNO₂Tz (+0.78 T), and c) CuNO₂Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

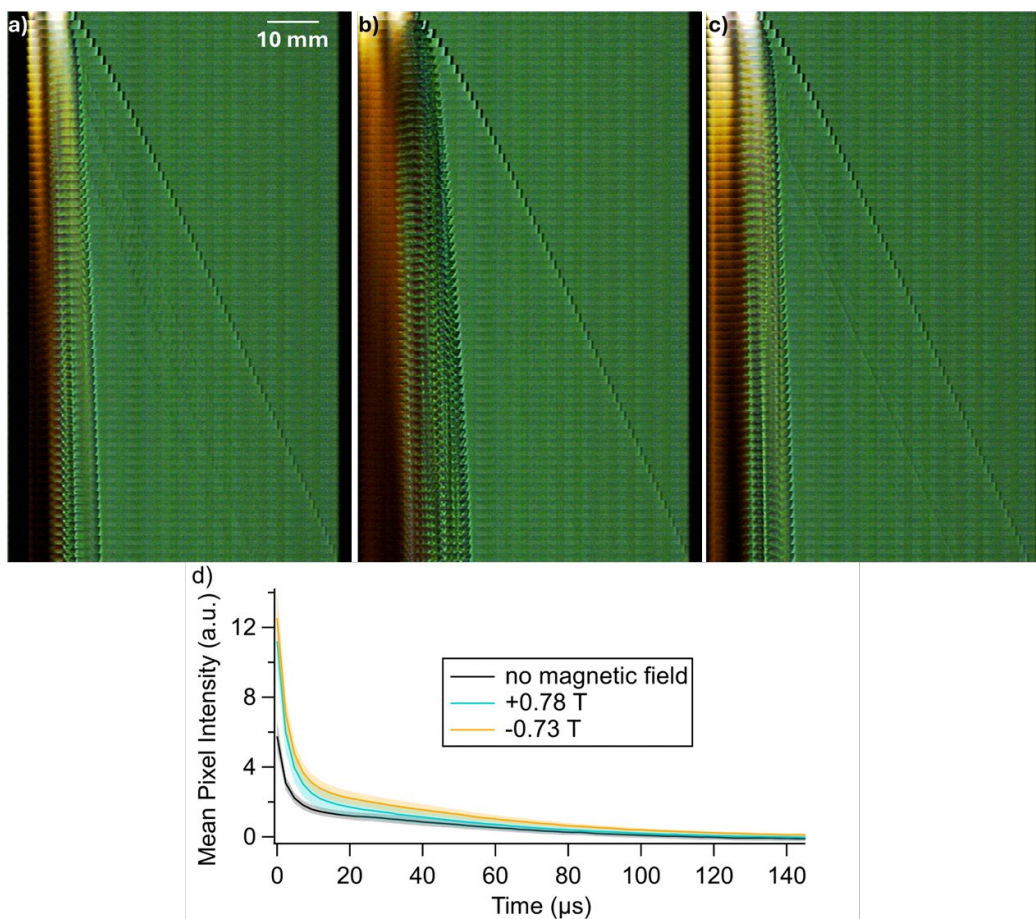


Figure A-3. Snapshots from representative high-speed schlieren videos of the a) CoNO₂Tz, b) CoNO₂Tz (+0.78 T), and c) CoNO₂Tz (−0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

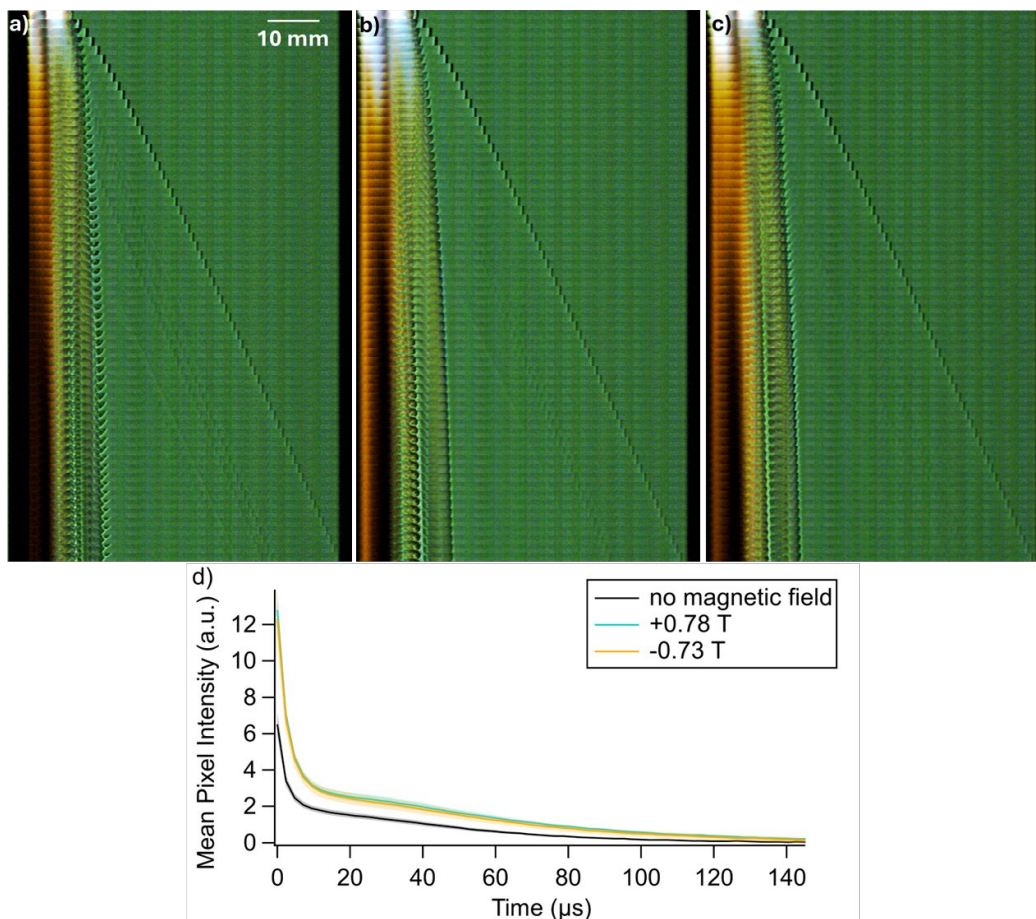


Figure A-4. Snapshots from representative high-speed schlieren videos of the a) FeNO₂Tz, b) FeNO₂Tz (+0.78 T), and c) FeNO₂Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

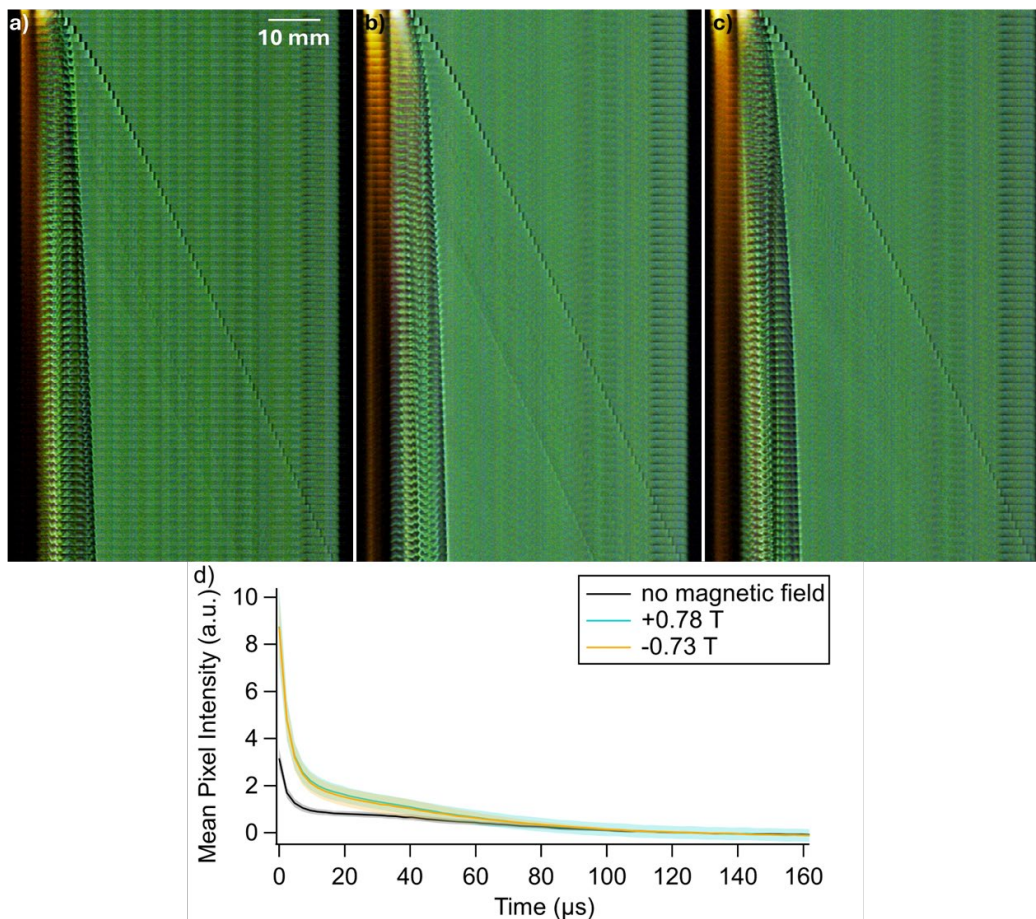


Figure A-5. Snapshots from representative high-speed schlieren videos of the a) NiNO_2Tz , b) NiNO_2Tz ($+0.78\text{ T}$), and c) NiNO_2Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

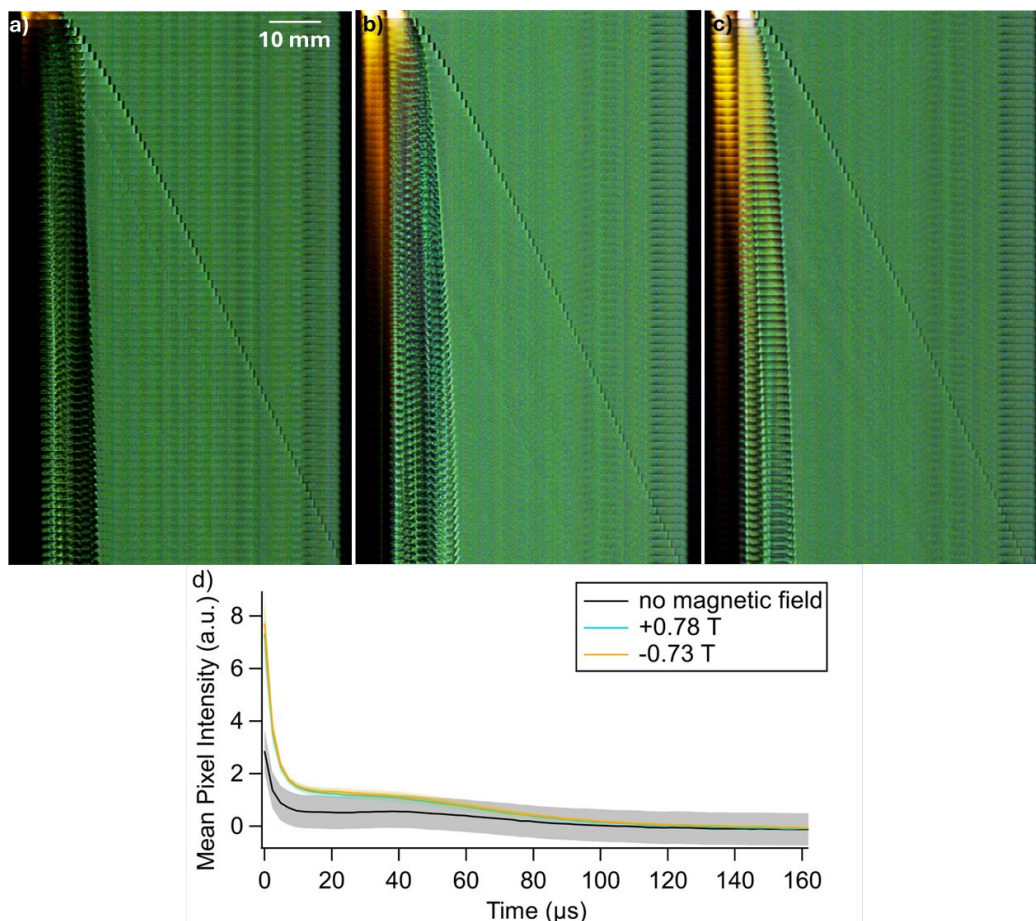


Figure A-6. Snapshots from representative high-speed schlieren videos of the a) ZnNO₂Tz, b) ZnNO₂Tz (+0.78 T), and c) ZnNO₂Tz (-0.73 T); d) the mean pixel intensities for all replicate videos were extracted and the average values are shown along with the 95% CIs.

Appendix B. Direct Comparison of Emission Spectra from each Time Regime

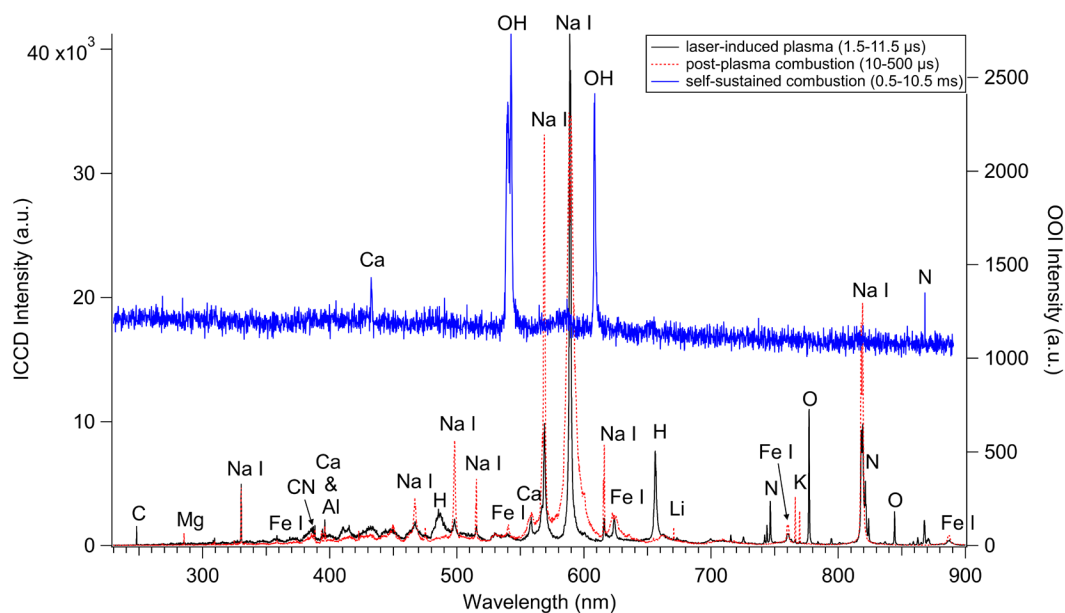


Figure B-1. Emission spectra for the NaNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

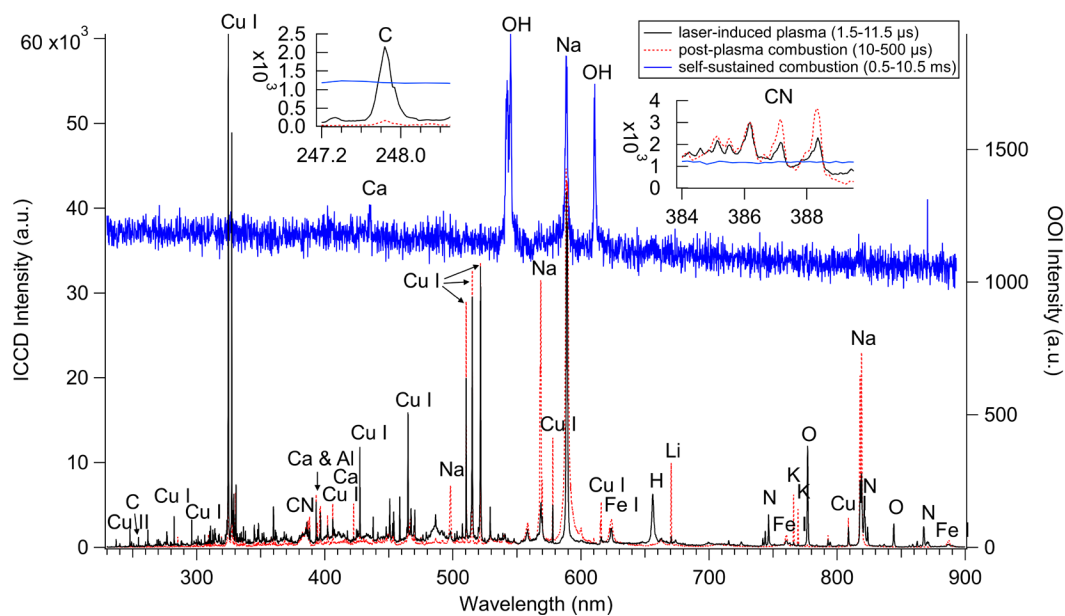


Figure B-2. Emission spectra for the CuNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

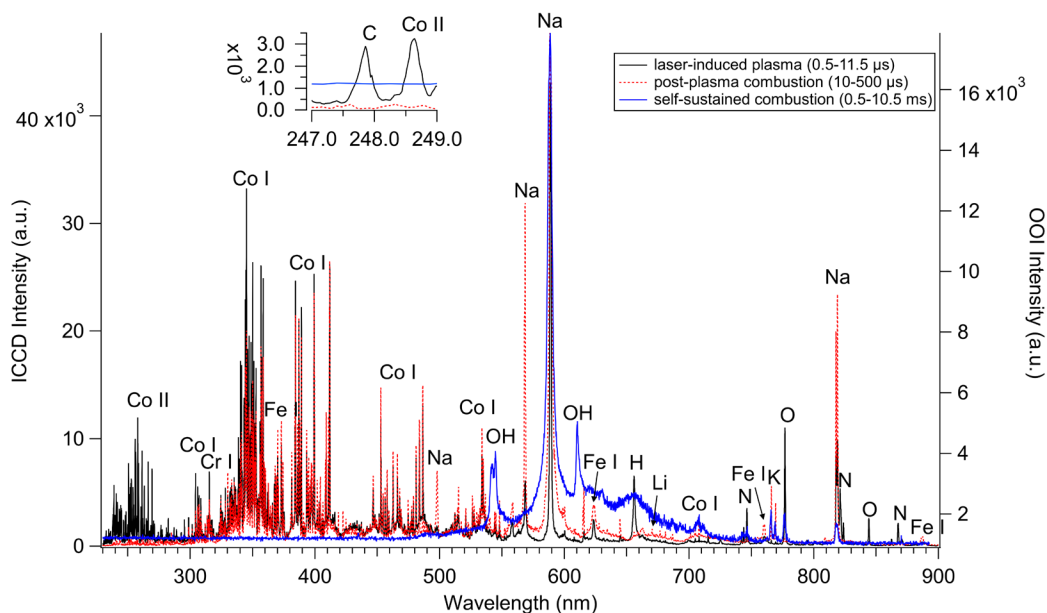


Figure B-3. Emission spectra for the CoNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

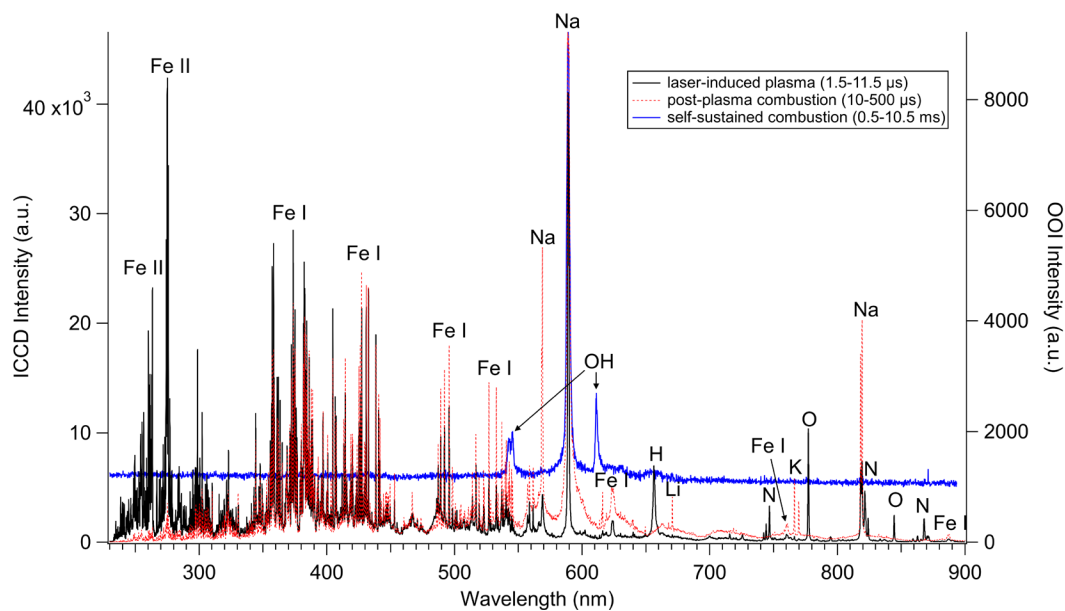


Figure B-4. Emission spectra for the FeNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

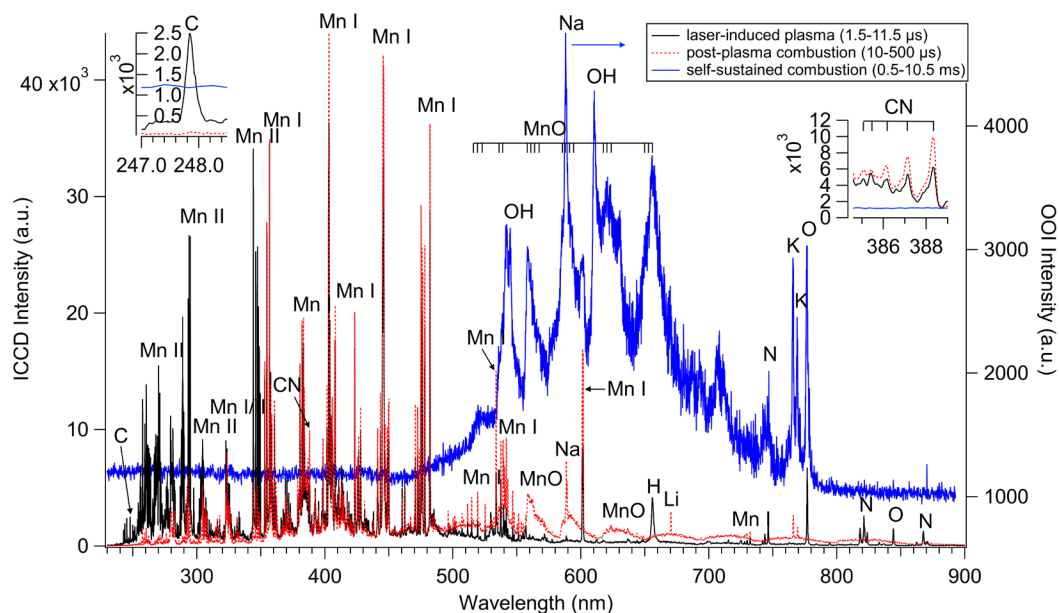


Figure B-5. Emission spectra for the Mn12 sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

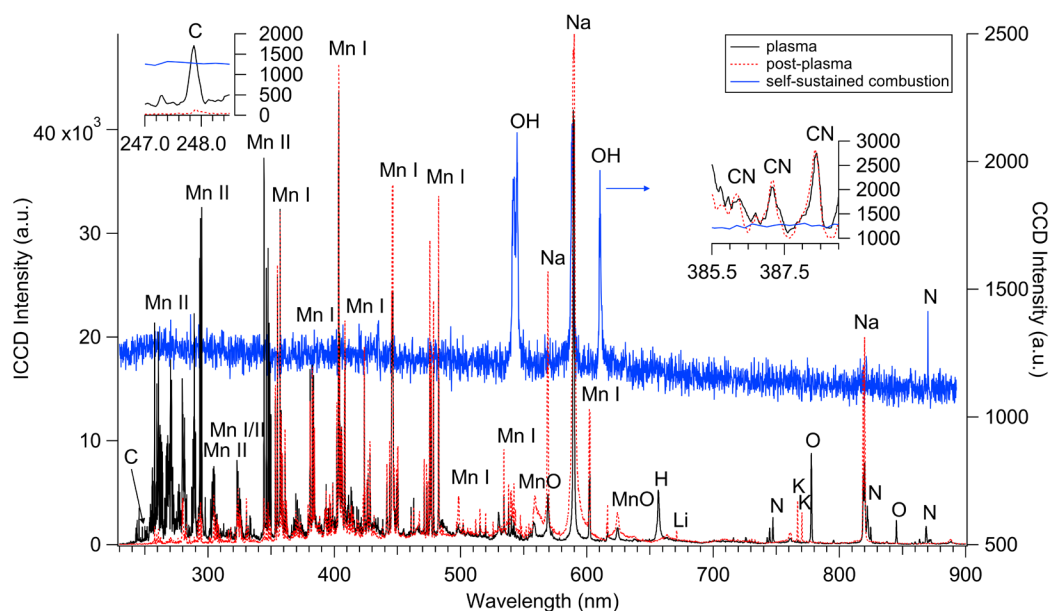


Figure B-6. Emission spectra for the MnNO₂Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

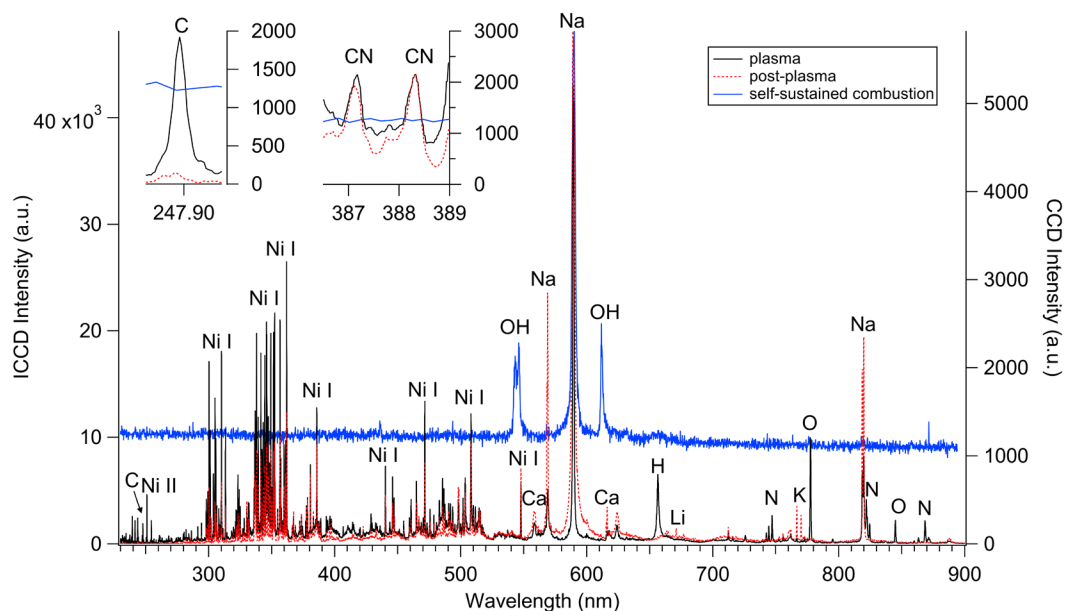


Figure B-7. Emission spectra for the NiNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

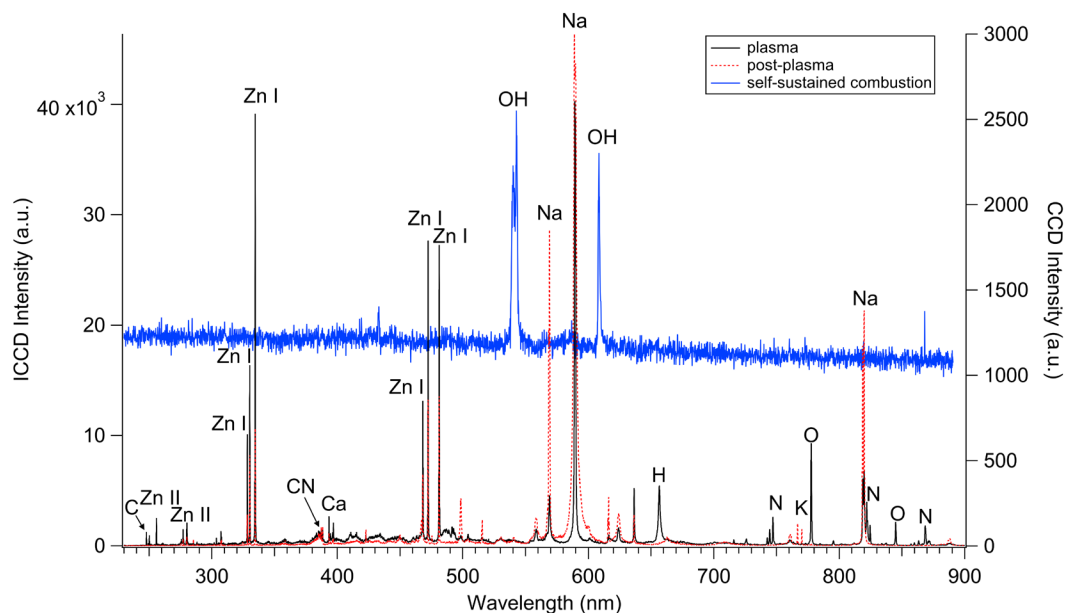


Figure B-8. Emission spectra for the ZnNO_2Tz sample during the laser-induced plasma (black), post-plasma combustion (red), and self-sustained combustion reactions (blue, right axis).

Appendix C. Additional Laser-Induced Plasma Spectral Data

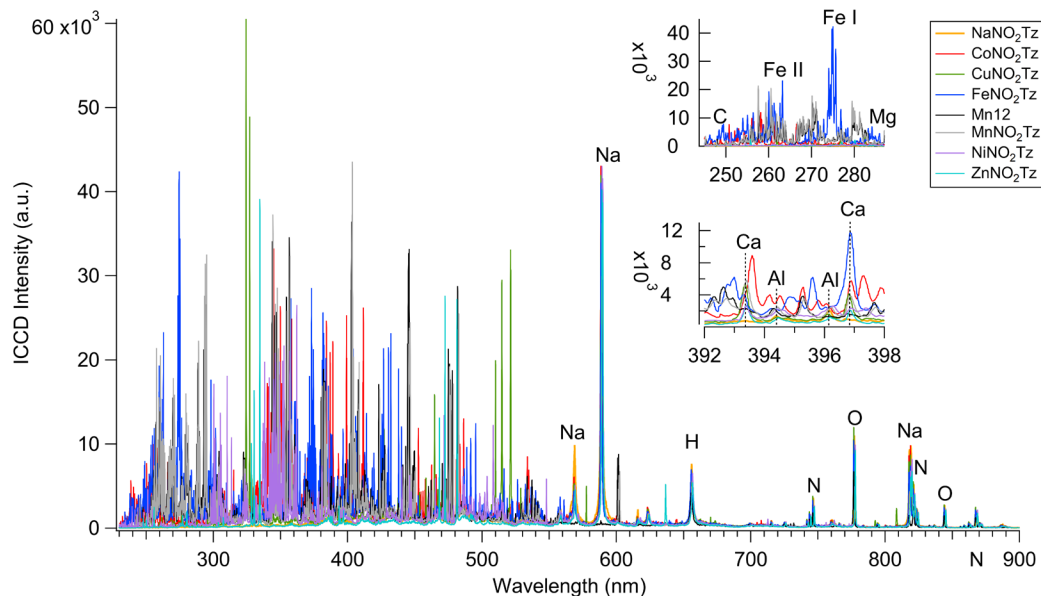


Figure C-1. Laser-induced plasma emission spectra for all samples with no magnetic field.

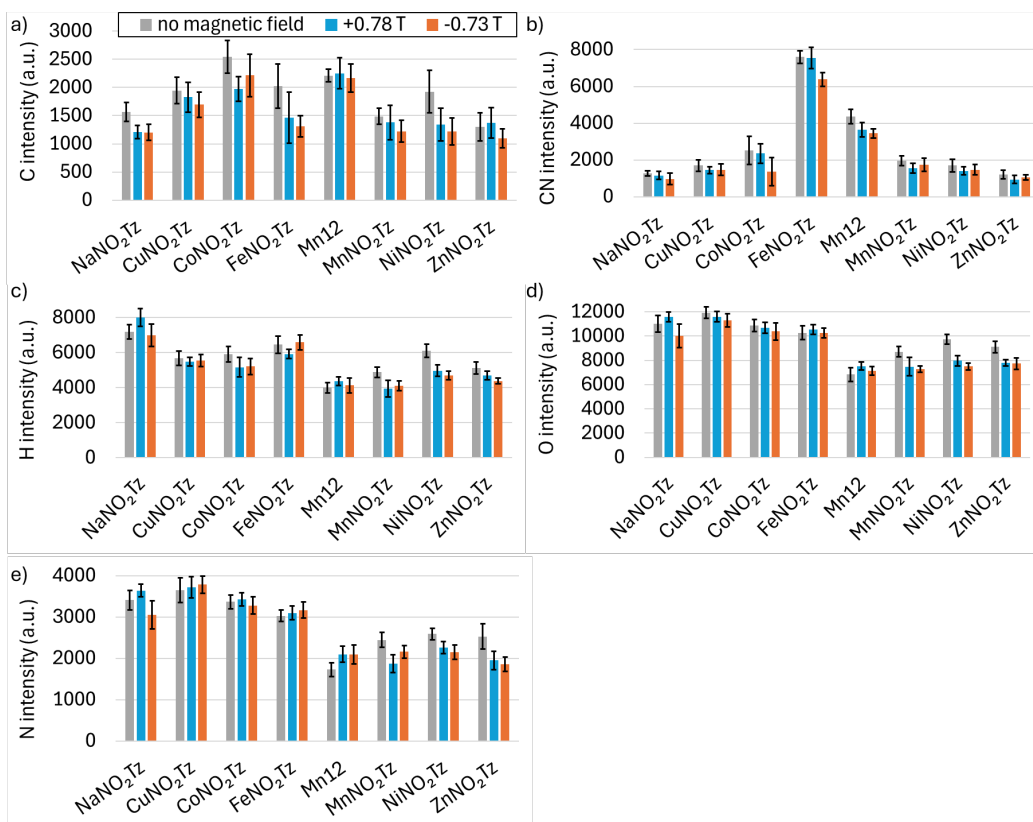


Figure C-2. Background-corrected peak emission intensities for a) C, b) CN, c) H, d) O, and e) N from the laser-induced plasma.

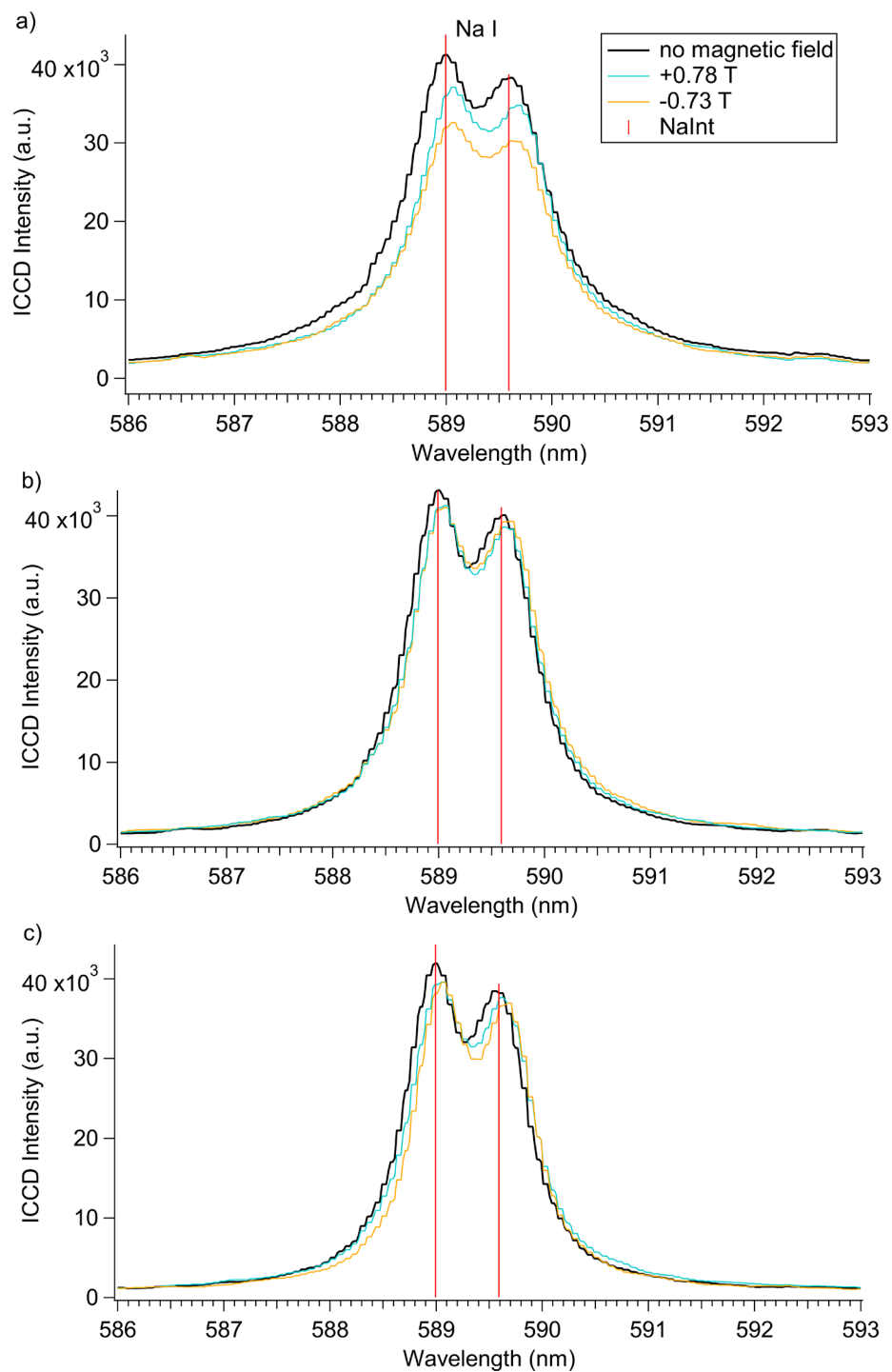


Figure C-3. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the laser-induced plasma.

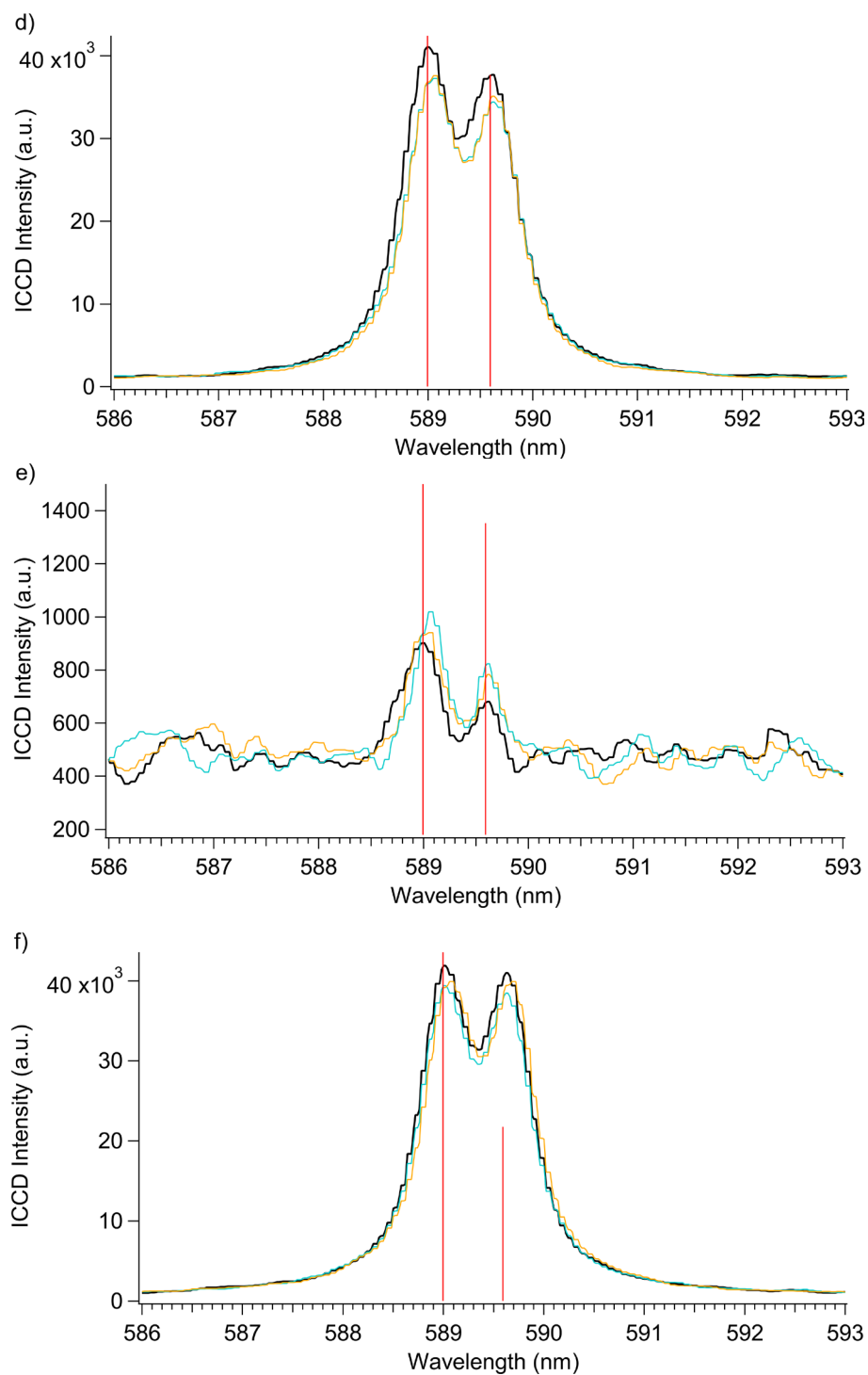


Figure C-3. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the laser-induced plasma (continued).

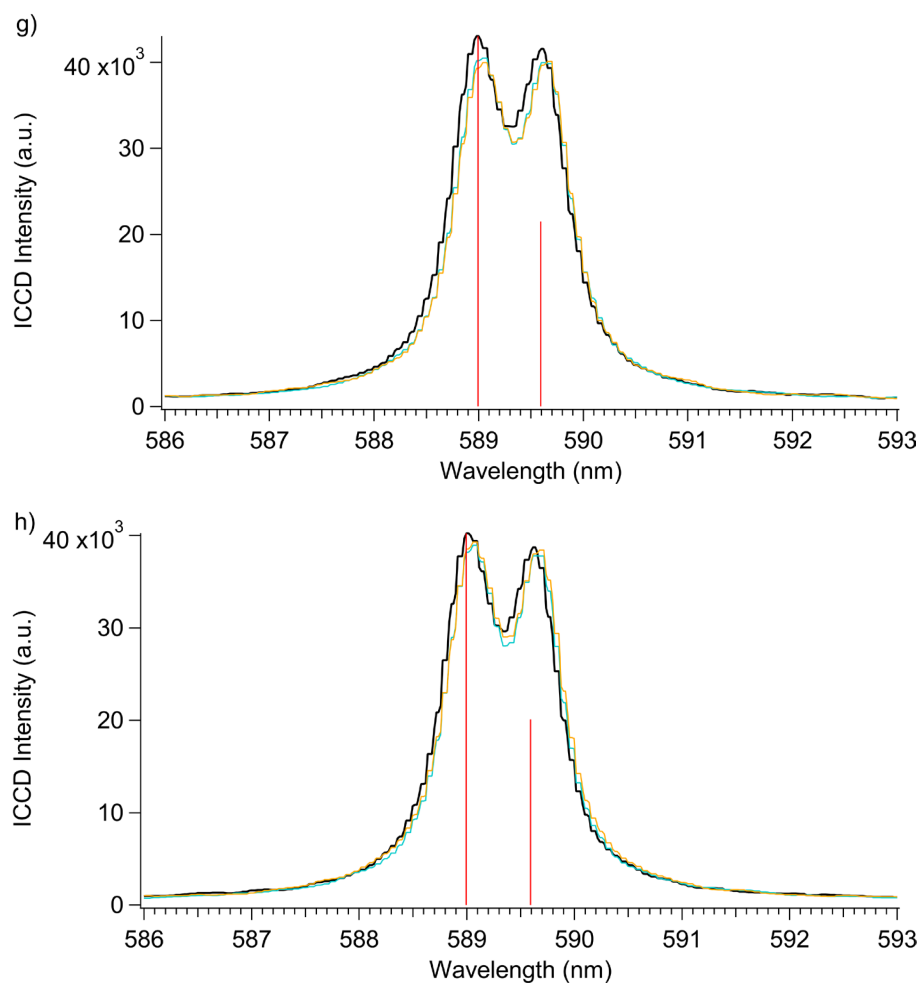


Figure C-3. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the laser-induced plasma (continued).

Appendix D. Additional Post-Plasma Combustion Spectral Data

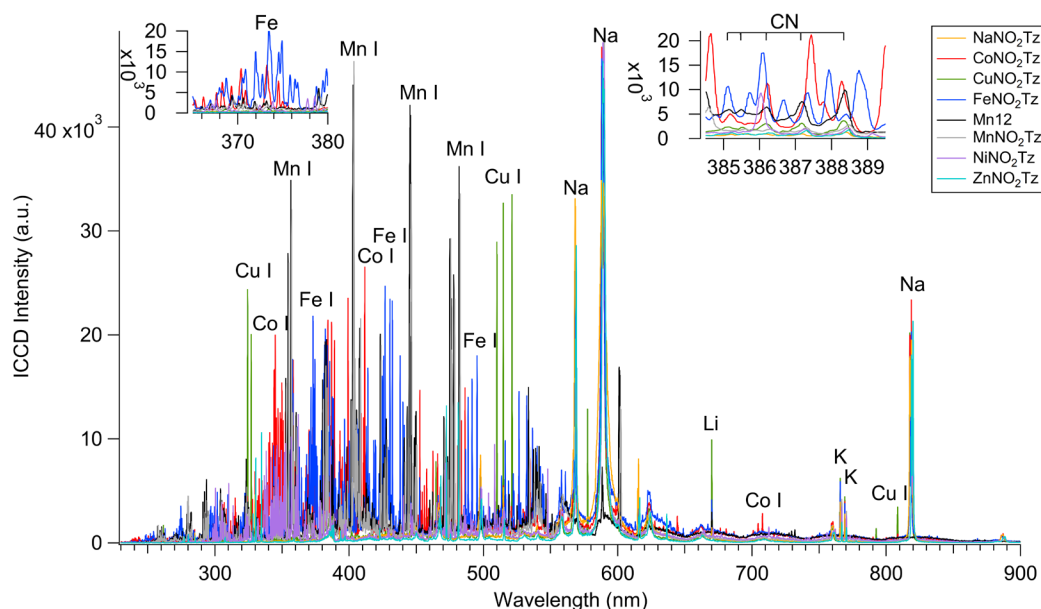


Figure D-1. Post-plasma combustion spectra for all samples with no magnetic field.

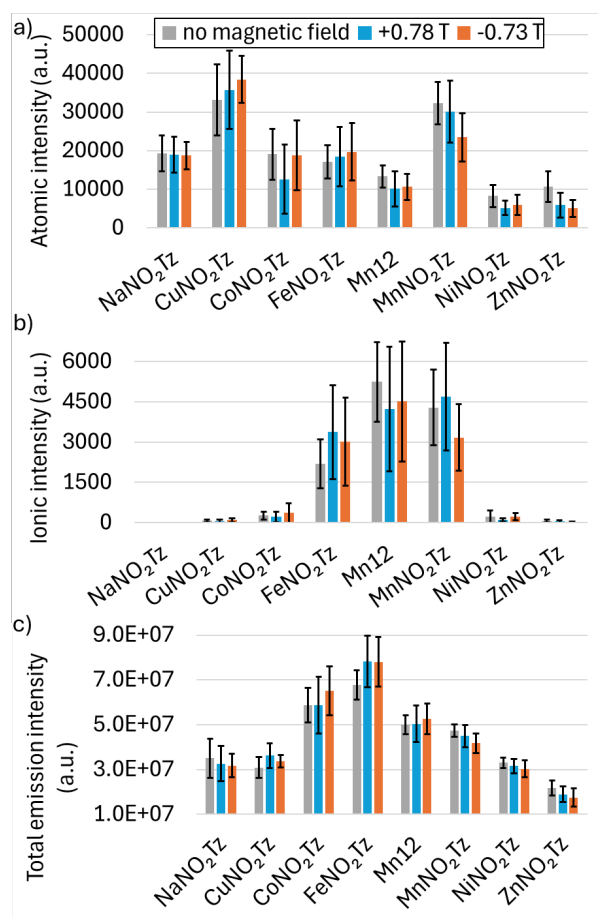


Figure D-2. a) Atomic, b) ionic, and c) total post-plasma combustion emission intensities for all samples with or without the magnetic field.

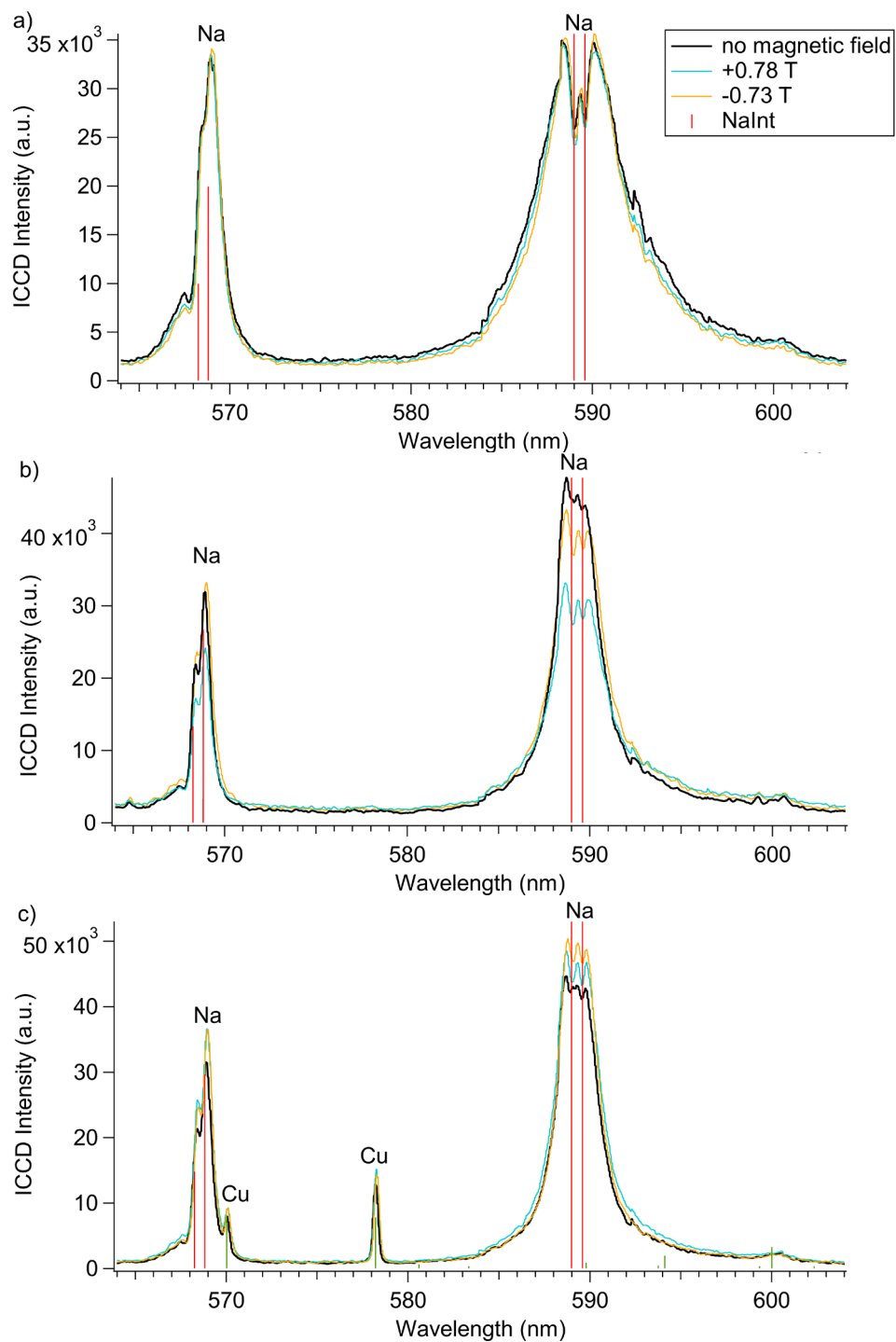


Figure D-3. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the post-plasma combustion.

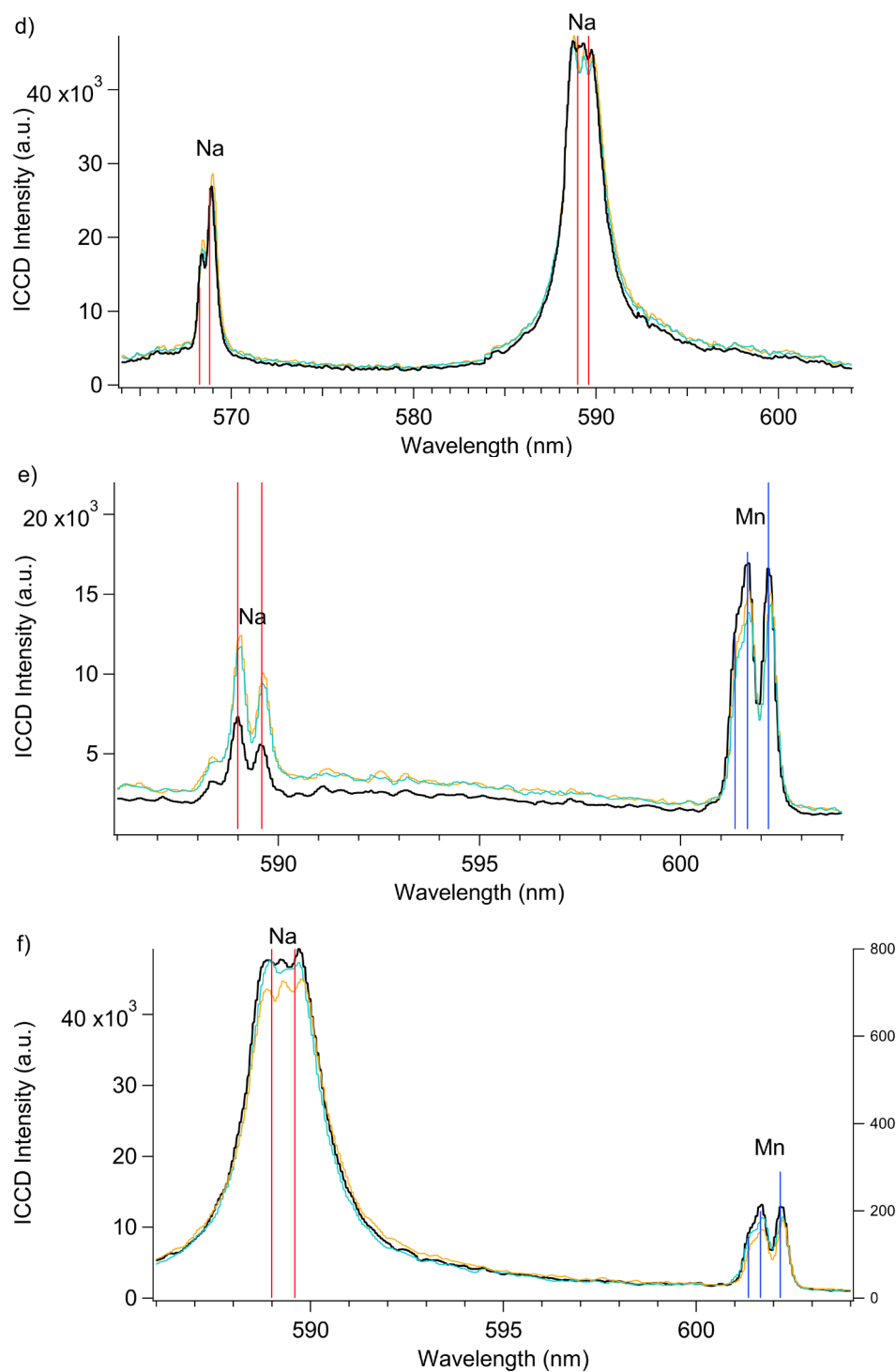


Figure D-3. Red-shift of the Na line for a) NaNO_2Tz , b) CoNO_2Tz , c) CuNO_2Tz , d) FeNO_2Tz , e) Mn12 , f) MnNO_2Tz , g) NiNO_2Tz , and h) ZnNO_2Tz in the presence of an external magnetic field during the post-plasma combustion (continued).

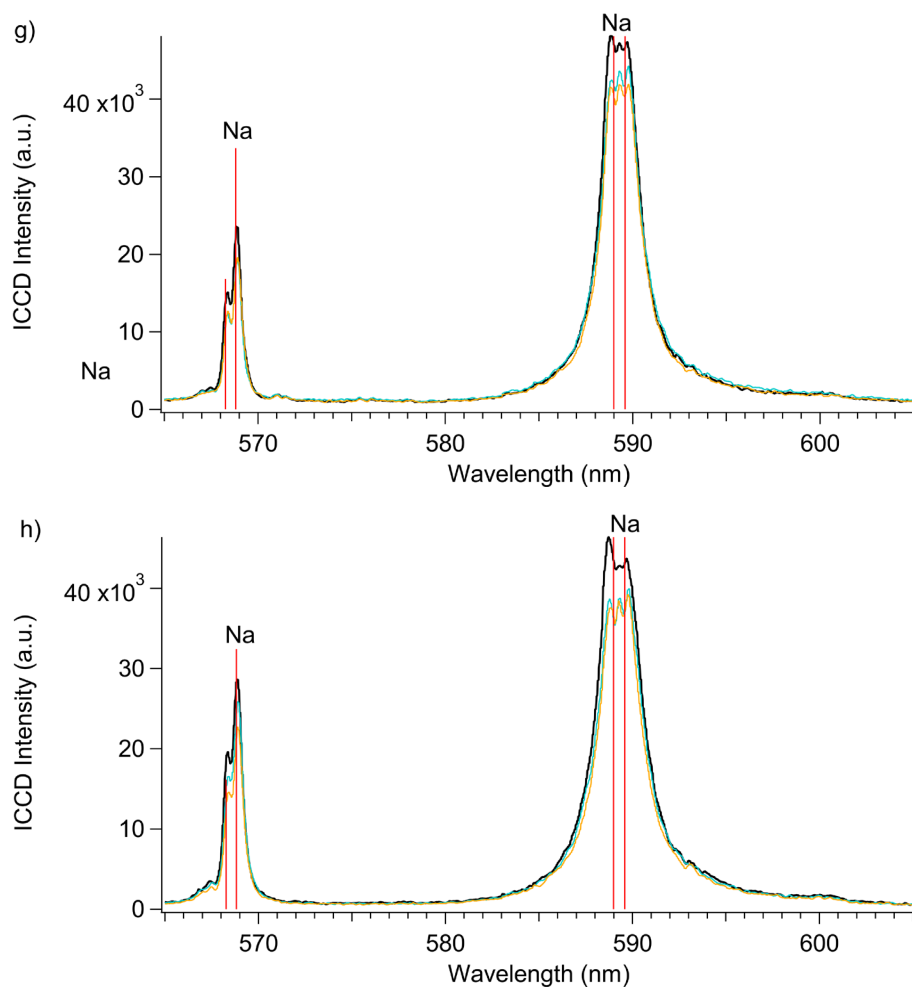


Figure D-3. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the post-plasma combustion (continued).

Appendix E. Additional Self-Sustained Combustion Spectral Data

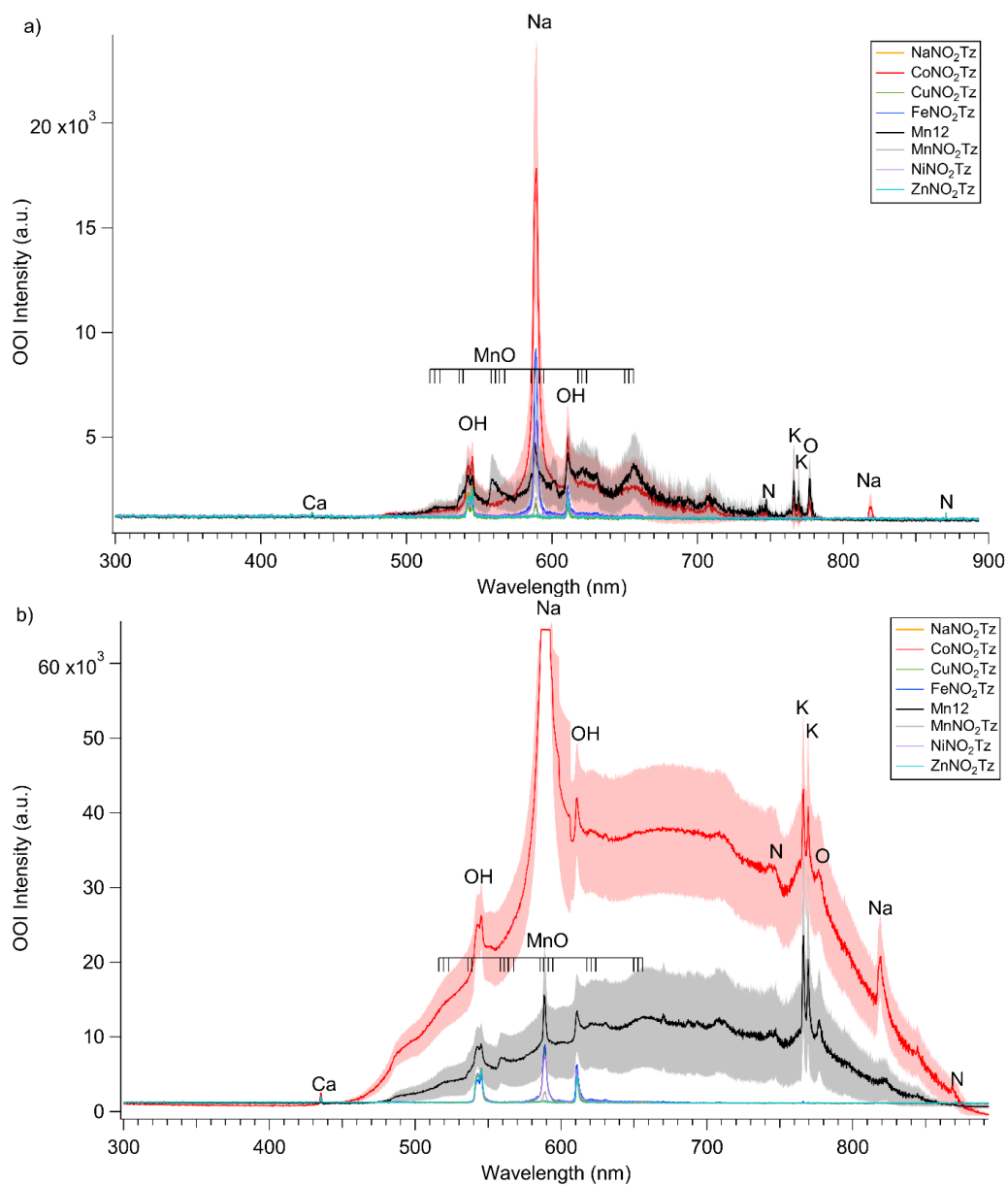


Figure E-1. Self-sustained combustion emission spectra for all samples with a) no magnetic field, b) +0.78 T, or c) -0.73 T. Error bars (95% CI) are shown for CoNO_2Tz and Mn12 .

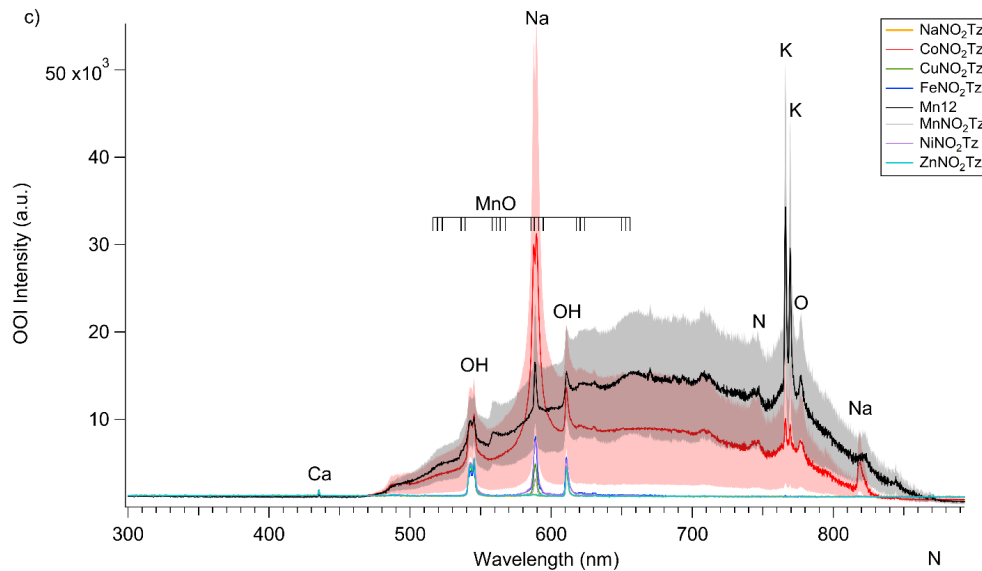


Figure E-1. Self-sustained combustion emission spectra for all samples with a) no magnetic field, b) $+0.78$ T, or c) -0.73 T. Error bars (95% CI) are shown for CoNO₂Tz and Mn12 (continued).

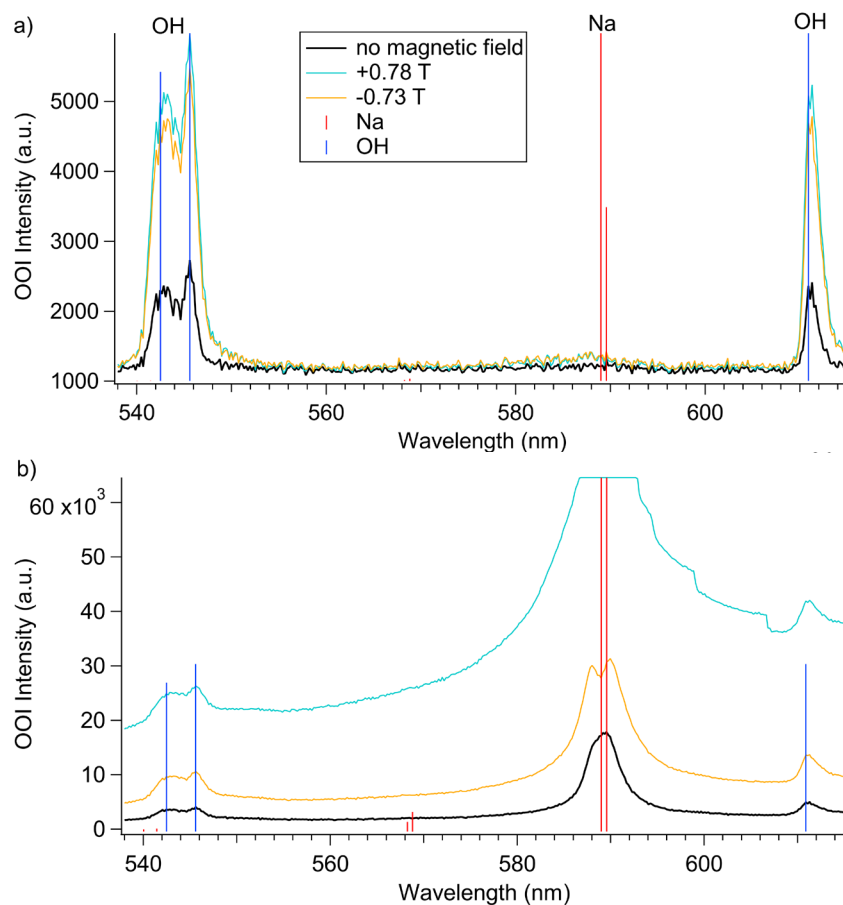


Figure E-2. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the self-sustained combustion reactions.

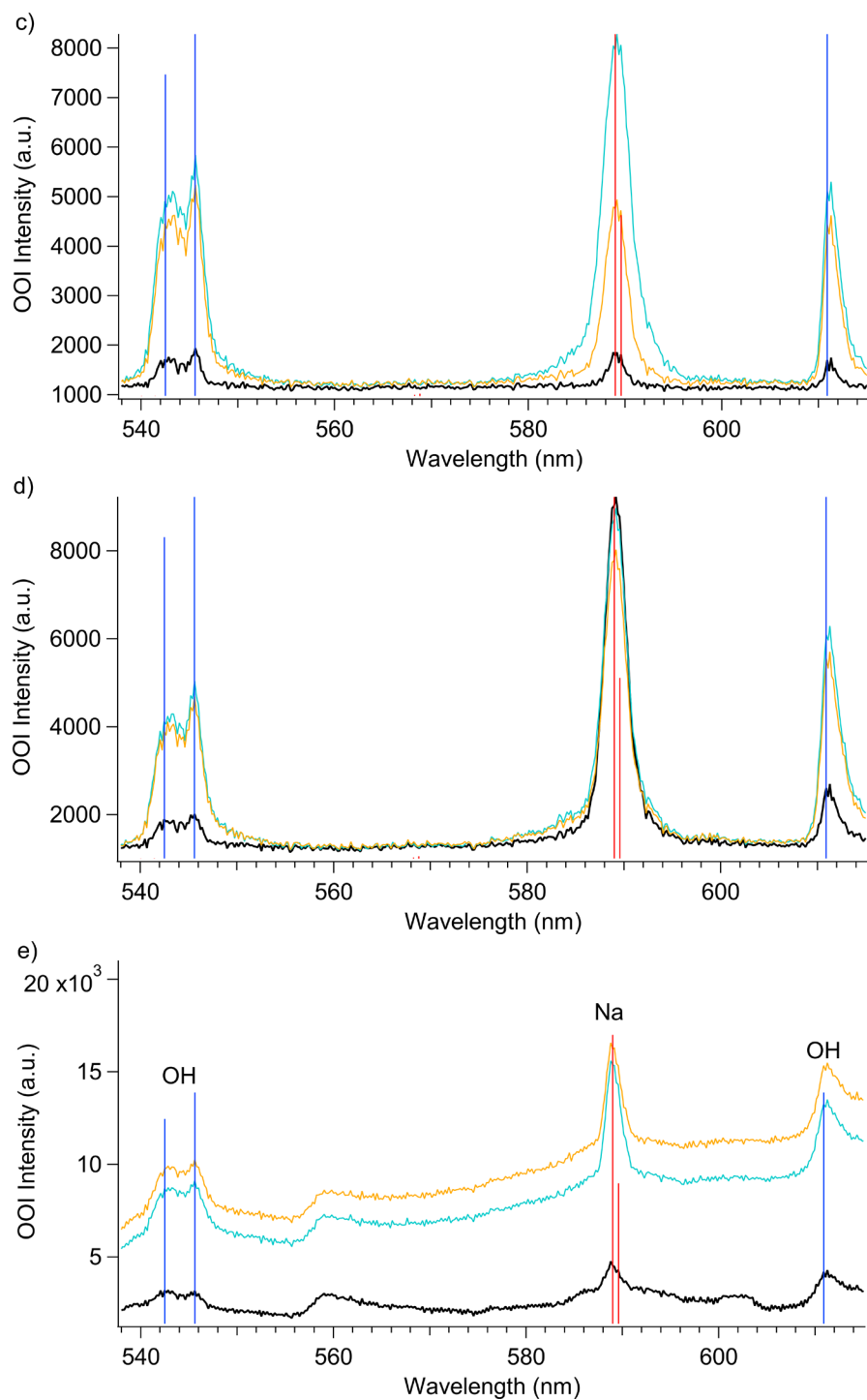


Figure E-2. Red-shift of the Na line for a) NaNO_2Tz , b) CoNO_2Tz , c) CuNO_2Tz , d) FeNO_2Tz , e) Mn12 , f) MnNO_2Tz , g) NiNO_2Tz , and h) ZnNO_2Tz in the presence of an external magnetic field during the self-sustained combustion reactions (continued).

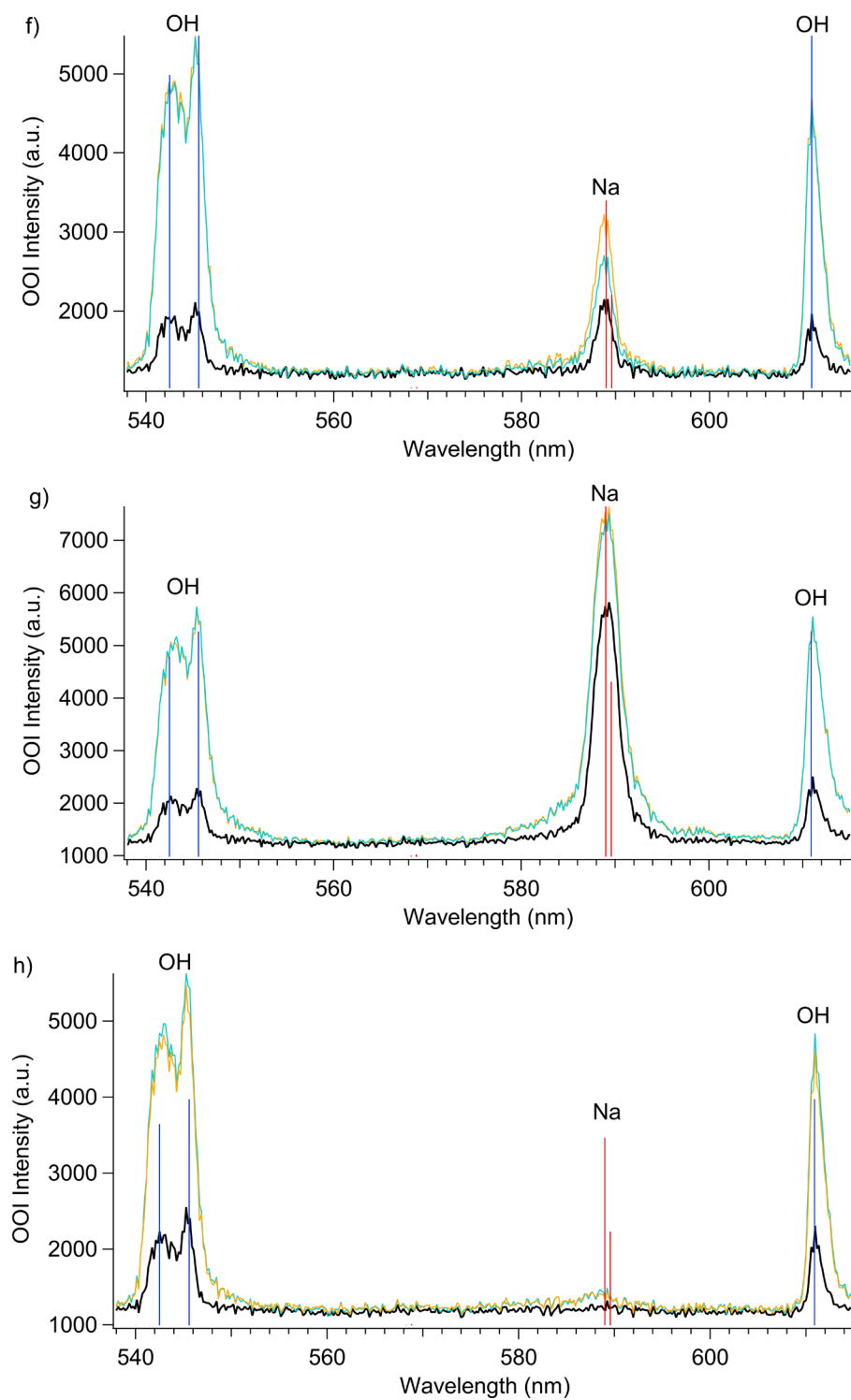


Figure E-2. Red-shift of the Na line for a) NaNO₂Tz, b) CoNO₂Tz, c) CuNO₂Tz, d) FeNO₂Tz, e) Mn12, f) MnNO₂Tz, g) NiNO₂Tz, and h) ZnNO₂Tz in the presence of an external magnetic field during the self-sustained combustion reactions (continued).

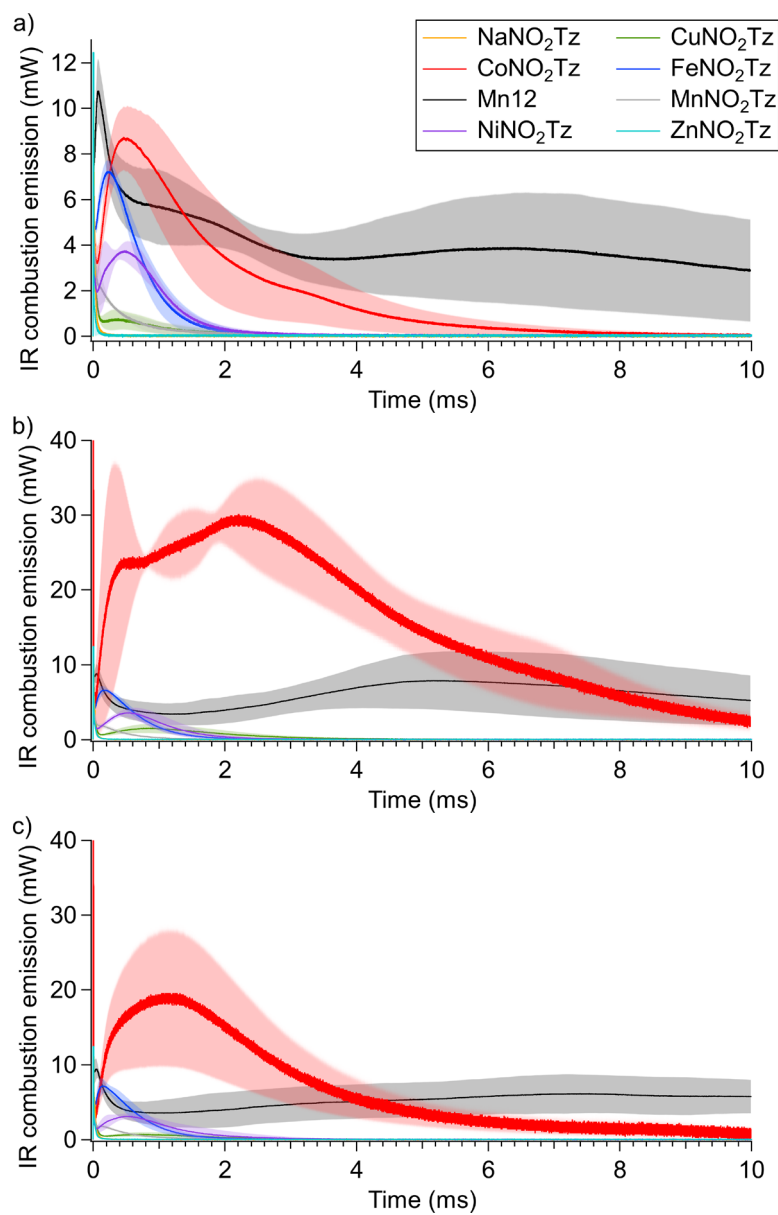


Figure E-3. Time-resolved self-sustained combustion emission integrated over the IR region for all samples with a) no magnetic field, b) +0.78 T, and c) -0.73 T. Shaded regions are 95% CIs.

List of Symbols, Abbreviations, and Acronyms

Al	Aluminum
ANOVA	Analysis of Variance
ARL	Army Research Laboratory
B	Magnetic-flux Density
B-LASEM	Magnetic-field-enhanced Laser-induced Air Shock from Energetic Materials
C	Carbon
Ca	Calcium
CCD	Charge-Coupled Device
CI	Confidence Interval
CN	Cyano Radical
Co	Cobalt
Cr	Chromium
Cu	Copper
DDG	Digital Delay Generator
DEVCOM	U.S. Army Combat Capabilities Development Command
DNOBz	3,6-Dinitrobenzoate
Fe	Iron
H	Hydrogen
ICCD	Intensified Charge-Coupled Device
IR	Infrared
K	Potassium
LASEM	Laser-induced Air Shock from Energetic Materials
Li	Lithium
Mg	Magnesium
Mn	Manganese

MnO	Manganese ^{II} Oxide
N	Nitrogen
Na	Sodium
Ni	Nickel
NO ₂ Tz	Nitrotetrazole
O	Oxygen
OH	Hydroxyl Radical
RGB	Red–Green–Blue
TNT	Trinitrotoluene
Zn	Zinc

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